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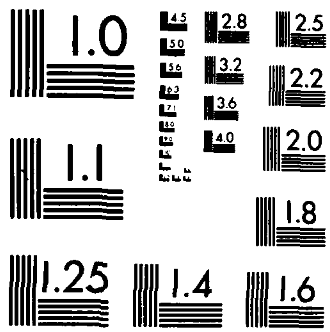

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Final report summarizes the results of 35 months research on the determination of (a) the dislocation densities of both edge and screw components using two positron annihilation techniques, and (b) the density of hydrogen traps by analysis of electrolytic permeation measurements. The dislocation densities of single crystal iron specimens deformed in three different manners were in good agreement with dislocation densities obtained by etch-pit and TEM methods on the same specimens as well as being bracketed by three earlier TEM investigations. ...2/.

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20. Abstract (contd.)

The positron lifetimes were 114 psec. in the trap-free bulk, 165 in an edge and 142 in a screw dislocation. The first two of these values are in good agreement with values obtained by several previous authors using poorly characterized specimens. It had been assumed that positron trapping in a screw dislocation was very unlikely so the value for the latter type of dislocation is an important result.

The binding energies of a hydrogen atom to an edge was deducted to be 37.4 and to a screw is 25.6 kJ/mole. The density of hydrogen at different strains was in good agreement with the number of dislocation traps. The occupancy of trapping sites along the dislocation was approximately 20 percent.

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FINAL REPORT

EFFECT OF INTERSTITIALS ON THE TRAPPING OF HYDROGEN IN HIGH PURITY IRON AS DETERMINED BY POSITRON ANNIHILATION AND TRANSMISSION ELECTRON MICROSCOPY

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Section I

OVERVIEW OF EXPERIMENTAL WORK

Most of the original objectives of this program were achieved: namely, it was established the occupation of traps by hydrogen atoms (or ions) could not only be detected but could be measured quantitatively by positron annihilation. One achievement beyond expectation was that the densities of edge and screw dislocations could be measured nondestructively. The permeation of hydrogen through deformed iron specimens was studied electrochemically and the density of hydrogen traps was determined as a function of strain.

The specific positron trapping rates were determined for the first time for edge and screw dislocations in Fe. Each has an identifying lifetime; namely, 165 and 142 psec respectively.

The position adopted by many in the field was that trapping by even an edge dislocation was not possible. Even though no quantitative support has been offered in the last twenty years, the position of Cotterill and Doyama[1] that trapping only occurs in jogs, has been widely accepted.

The present data and analyses strongly support trapping by dislocations, not only edge, but screw dislocations as well. The density of each for various strain levels obtained by positron annihilation measurements agree well with TEM and etch-pit determinations carried out with the same specimens. In addition, these measured densities lie with the bracket; i.e., in the range of three previous TEM studies[2,3,4]

Having electrochemically measured the transport of hydrogen through the specimens which had already been measured with positrons, we could obtain both the average concentration of hydrogen C_{av} and the density of dislocation traps N_v. The occupation ratio could be obtained from these two numbers; it ranged from 0.05 to 0.14. The reduction of the trapping rate, i.e., the fraction of positrons annihilating in an edge or screw dislocation trap when hydrogen was introduced, was another measure of occupancy; these ranged from 0.10 to 0.27. These can be compared with the value which Gibala[5] obtained in connection with internal-friction measurements, namely 0.25. The reduction in the positron annihilation was caused by the partially ionized hydrogen atoms blocking the positron traps. This is evidence that the occupation of the "irreversible" hydrogen traps was low as we deduced from the analysis of the transport of hydrogen through the deformed metal.

The same experimental data was treated by the several models in the literature for permeation of hydrogen through defect-containing iron[6,7,8] and a spread of the calculated trap densities was at least a factor of 8 for each strain level. This probably resulted from the various binding energies assumed by the those authors - seven different values ranging from 19.2 to 59.9 kJ/mole have been reported. The equation from the model of Choi[7] which did not directly involve a binding energy gave trapping densities in very good with those determined by positron annihilation. Oriani's equation

$$E_b = -RT \ln [N_T (D_L/D_a - 1)^{-1}]$$

was used to determine the binding energies from (a) bent specimens, (b) cold rolled and (c) low-temperature deformed specimens.

Two binding energies for hydrogen were determined in this way, namely 37.4 kJ/mole for edge and 25.6 for screw dislocations. These values are in good agreement with other studies of hydrogen trapping.

The strong correlation between positron (dislocation) traps and the hydrogen traps indicates that the two different "particles" share a common dislocation-related trap.

It has been shown in this work that more extensive charging, i.e., a hundred-fold increase in hydrogen charging of slightly deformed specimens does not lead to a new positron lifetime and that on removal of hydrogen *in vacuo* at room temperature the trapping rate κ_t returns to its pre-charging value. Thus no generation or damage by the hydrogen is indicated. The idea of trapping in a dislocation is strengthened

by these data.

With the hundred-fold increase in the charge density of hydrogen, an additional lifetime occurred which was longer than any the authors had previously observed i.e., > 165 psec. The single-lifetime fitting gave 180 and 190 psec. This could be explained by the formation of vacancy clusters. The trapping rate for this lifetime was reduced by the presence of hydrogen but it failed to return all the way to the pre-charging value. On removal of the hydrogen an interpretation is that some damage was introduced by more extensive charging and that the hydrogen was then more difficult to remove from the new trap by pumping.

It has been shown that there are insufficient jogs (hundreds of Angstroms long) visible in the authors' TEM pictures to account for the direct trapping into dislocation traps as determined by positron annihilation. Nevertheless, the experiments to date do not exclude the possibility that positrons are predominantly annihilated in jogs if the assumption of possible "pipe" diffusion is included. Nevertheless, the authors are not aware of any studies which show enhanced diffusion of interstitials (in contrast to substitutional impurities) down dislocation in BCC metals. There may also be significantly more jogs of almost atomic dimensions in the deformed specimens[9] which are not resolved by the TEM instrument used. This question is being investigated theoretically.

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Section I A

Ph. D. Research Supported
Yong-Ki Park March 1985

Title: Determinstion By Positron Annihilation And Electrolytic Hydrogen Permeation Of The Dislocation and Hydrogen Trap Densities In Annealed And Deformed Iron Single Crystals.

Post-doctoral Fellows Supported
Wilfred Alsem
William Friess

Undergraduate Assistants Supported
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Aaron Parks

Section II
OVERVIEW OF THEORETICAL WORK

The theoretical work dealing with trapping of positrons in vacancies and vacancy clusters has mainly been carried out by the Prof. Nieminen and Puska[T1] in Finland and Chakraborty[T2] at Argonne. There is essentially no work which deals realistically with dislocations. This will be undertaken in the near future.

Nickel oxide was studied experimentally because, as is well known in connection with oxidation of metals, one can control the number of vacancies by treatment in various oxidizing or reducing atmospheres. Experimental lifetime of a positron in bulk 149.5 psec agreed remarkably well with the theoreticaal value 150 psec. calculated from the band structure of positrons[3] in Nickel Oxide. When this paper was presented at the 6th International Conference on Positron Annihilation in 1982, it was criticized because of the approximate method used to treat the correlation between the positron and the electrons. Since that time we have made efforts to improve this situation but they have been time-consuming.

In the absence of good theoretical or experimental studies on the correlation of core electrons in metallic solids or in ionic compounds, it is difficult to assess whether the any proposed treatment does adequately account for the effects of correlation. In as much as the binding of the positron to a simple molecule does not involve exchange forces but only electrostatic and correlation forces, calculation of the

incipient binding and/or attachment affords an opportunity for direct comparison with experiments. Thus, there is a real advantage to studying gaseous molecules before ionic solids and metals. There are a number of experimentalists measuring now or tooling up to measure the positron cross section of slightly more complex atoms and molecules as a function of energy. The vapor of the alkali metals is a prime candidate. The formation of simple molecules involving an atom plus a positron is a more tractable case than either ionic solids such as alkali halides or metalsa are. Stepping stones are needed to evaluate whether the theoretical method is adequate and whether one can unravel the various contributions before one tackles a really large problem.

As an aside, the experimental measurments on the alkali halides need to be redone with more carefully characterized specimens.

One merit of this theoretical work is that one can avoid calculating the exchange energy of the positron since it is distinguishable from the other Fermions, i.e., electrons in the gas or solid. Thus in addition to the electrostatic potential which is readily handled there is only a correlation term to be evaluated and compared with experimental results - i.e., there is no uncertainity due to approximations introduced by the choice of the exchange potential.

Several papers have been submitted in this area and the ensuing reviews have questioned how large is the electron-electron correlation term when it is also done by second-order perturbation calculations since the electron cloud will be

influenced by the presence of the positron? The question was not posed as an insurmountable barrier to publishing by the reviewers. We have chosen voluntarily not to publish the early results until the problem could be treated more thoroughly. Without going into details, this has necessitated a complete overhaul of the codes; otherwise the amount of machine time involved would be almost prohibitive. The computer programs have now been written for significant memory and speed enhancements on the Super-mini computer and array processor at Michigan Technological University and tested with lithium at Urbana prior to full scale use at MTU.

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Section II - PRESENTATIONS

"Positron Annihilation and Electrolytic Permeation Study of Iron Single Crystals"
Authors: *Yong-Ki Park,* W. Alsem, C. G. Park, James T. Waber C. L. Snead, Jr. and K. G. Lynn
Place: TMS-AIME Meeting, Atlanta, Ga. March 1983

"Correlation in Exotic Molecules using Second-Order Perturbation Theory for Assessment"
Authors: *James T. Waber,* Asok Ray, A. B. Kunz and R. Weidman
Place: Eight Canadian Symposium on Theoretical Chemistry, Halifax, Nova Scotia, Canada, August 1983.

"Dislocations and Hydrogen Embrittlement in Iron Single

Crystals"
Authors: *James T. Waber* and Yong- Ki Park
Place: First Review of Research Conference on Materials Research, Newly Formed Chicago Section, TMS-AIME, Chicago, Il Sept. 1983

"Characterization and Behaviour of Materials with Sub-Micron Dimensions"
Author: *James T. Waber*
Place Joint ASM-TMS Symposium, Philadelphia, Oct., 1983

"Study of Dislocations in Alpha Iron and the Effect of Hydrogen on Positron Trapping"
Authors: *James T. Waber,* Yong-Ki Park, C. L. Snead, Jr. W. Lanford and K. G. Lynn
Place: Materials Research Society, Boston, MA Nov. 1983.

"Determination of Edge and Screw Dislocations in Iron Crystals using Positron Annihilation"
Authors: *Yong-Ki Park,* James T. Waber, C. L. Snead, Jr. and K. G. Lynn
Place: TMS-AIME Annual Meeting Los Angeles, CA Feb. 1984.

"Determination of Edge and Screw Dislocations and the Effect of Hydrogen in Iron Single Crystals"
Author: *James T. Waber*
Place: Collquium, Michigan Technological University, Houghton, MI July 1984.

"Effect of Fermion Mass on the Formation of Bound States in Hydride-Like Exotic Molecules"
Authors: *James T. Waber,* A. B. Kunz and Neil Roeth
Place: American Symposium on Theoretical Chemistry, Boulder. CO Aug. 1984.

"Behavior of Materials with Sub-Micron Dimensions - The N=100 Atom Problem"
Author: *James T. Waber*
Place: DOE Panel on Theory ond Computer Simulation of Materials Structure and Imperfections, Houghton, MI Aug. 1984

"Methane Bubble Formation in Hydrogen Charged Fe-30 ppm C"
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An earlier draft of the positron manuscript was reviewed by Professor R. A. Oriani of the University of Minnesota; he made very useful comments. The technical assistance of W. Tremel at BNL is greatly appreciated.

A very important contribution to this work was the single crystals supplied by Professor M. Meshii. Purification of the specimens was carried out in a vacuum furnace made available to us by him. Large strap-shaped single crystals of high-purity iron were also provided by Professor J. T. M. de Hosson of the University of Groningen.

a positron) down the more open dislocation core would occur significantly more rapidly than through the unstrained lattice.

One can envision the saddle-point maxima acting as conventional diffusion barriers for positron diffusion which are larger along edge-dislocation core than they are in the undefected lattice, thereby providing possible traps ranging from deep to metastable. A similar argument could be offered for trapping and annihilation in the kinks of screw dislocations. The case for ultimate trapping of positron at jogs along edge dislocation (and kinks on screw) is not well established. It can be shown to be one plausible explanation.

CONCLUSIONS

The density of dislocations as determined by positron annihilation was found to be in good agreement with other independent measurements: namely, with etch-pit and TEM determinations. Positrons offer a nondestructive method for measuring the fraction of edge and screw components formed during deformation. In agreement with statements in the literature the density of screw dislocations generated by tensile deformation at 200 K was found to exceed that of edge dislocations. Trapping at jogs and kinks is plausible, but needs investigating to be demonstrated as an established mechanism.

ACKNOWLEDGMENTS

The assistance and counsel of Dr. Kelvin Lynn of Brookhaven National Laboratory and of Professor I. K. Mackenzie of Guelph University in the early stages of this work is gratefully acknowledged. Dr. Wilfred Alsem of Delft University was able to locate and correct an electronic flaw in the lifetime measuring system at Northwestern University. The authors are also indebted to Professor A. Vehanen and Dr. B. Nielsen for their very helpful discussions.

similar to that of a single vacancy, this concentration is too small by more than two orders of magnitude to account for the dislocation trapping observed herein. Direct trapping of (large) jogs can thus be ruled out.

Smedskjaer et al. [2] however, contend, within the assumptions of their model (for example, 0.01 eV binding energy) that with a jog density of one jog per 100-1000 Burgers vectors along the dislocaton, it is probable that most positrons that localize initially on the dislocation line will trap at the deeper jog trap before annihilation. We note that this jog density is comparable to that in the specimens investigated here. Thus, the Smedskjaer's analysis would suggest that the majority of the positrons that localized in (on) dislocation in these Fe specimens could annihilate from jogs.

The two key assumptions that enables this are: (a) "pipe" diffusion of positrons down the dislocation core funneling the localized positron toward jogs and (b) the jogs are deeper traps. The rate-limiting step in positron trapping then is not the trapping at the jog with its cross section determining the rate, but rather the step of trapping on the dislocation line itself [20].

One can, however, say that the annihilation [trapping] at the jogs is plausible but in no way proven. The crucial assumption of "pipe" diffusion of the positron along the dislocation core after localization is not established. The authors have not been able to locate references to independent evidence for "pipe" diffusion of interstitial atoms in bcc metals. The value for the activation energy for lattice diffusion of hydrogen near room temperature in iron is only 6.7 kJ/mole [20]. It seems unlikely, in view of the little lattice strain is involved, that diffusion of even a hydrogen atom (let alone

to the dislocation or something associated with one, such as a jog along an edge, or a kink along a screw dislocation, is left unanswered. Complementary measurements of the defect structure in the deformed specimens were employed to investigate this question.

More-direct evidence is the TEM pictures taken of (A) crystals strained in tension at 200 K and (B) crystals rolled at room temperature. The long parallel <111> screw dislocations in Type A crystal are presented in Figs. 3 and 4. Mixed-dislocation tangles were found in the cold-rolled crystals. In Fig. 5, the (total) dislocation densities found by positron annihilation in specimens rolled at 300 K are compared with the TEM results of Keh [6] and Yamashita et al. [16]. The authors' TEM determinations are also shown. While the errors shown on the positron lifetime results are large, they are properly compared with the probable errors shown in the TEM results of Ikeda [17] which span in some cases an order of magnitude in density.

The relatively straight form of the dislocations in Figs. 3 and 4 as well as other similar TEM pictures by the authors indicates that very little cross slip occurred during the deformation or even during the preparation of the thinned specimens for electron microscopy. Thus, there is little reason to believe that the number of jogs (or better large jogs [18]) was large enough to account for the measured number of traps.

In slightly compressed samples o. silicon iron, Low and Turkalo [19] measured a (large-) jog density correspondong to one jog per 2000 Burgers vectors for dislocations along <111>. If we associate this with the dislocation density found in this work, a concentration of jogs of 2.4×10^{-9} per Fe atom results. If a jog is assumed to have a trapping cross section for positron

a few psec of 114 for the bulk and the other within a few psec of 165 for an unidentified dislocation [3,11-13].

On analyzing data collected for single crystals cold-rolled at room temperature, the lifetime for a single trap was 154 psec. Vehanen [14] suggested that this conflict could be resolved if the value of 165 represented a mixture of 155 for a dislocation and 175 psec for monovacancies, since the latter could also be produced during deformation. On analyzing our data for cold-rolled specimens we obtained no statistical justification for retaining 175 psec. Our counterproposal took account of the complex stress condition during rolling which would produce both edge and screw components and predicted that a smaller lifetime between 165 and 114 might exist for a screw dislocation; that is, that 154 for a single trap could be separated into different fractions of positrons annihilating with lifetimes of 165 psec and a shorter time. In this way 142 psec was found.

A number of experiments were done to confirm the conclusion that 142 psec should be associated with screw dislocations. In addition to analyzing runs with increasing cold-working reductions in area at room temperature, (a) a series of well-annealed single crystals were strained in tension at 200 K and (b) iron whiskers were deformed in torsion at 300 K. These deformations are expected to yield predominantly screw dislocations. In agreement with the literature [15], the ratio of edge to screw components was larger than unity at low deformation at 200 K but rapidly decreased below 1 as more screw dislocations were produced at higher deformations. This trend is shown in Fig. 1. The lifetime of 142 psec was the dominant one for the twisted whiskers.

While a connection between the 165 and 142 psec t aps and leformation is shown by these experiments, the question of whether the trap is directly due

recommended by Shemenski et al. [7], etch pits were produced [8]. The density for different degrees of strain were determined from the TEM pictures of the replicas. The number of emergent dislocations is compared in Fig. 1 with the total number obtained with positron annihilation. It is reasonable that the values from etch pits might be smaller than those from the positron probe since not all dislocations intersect the surface.

It had not been anticipated at the outset of this research that one could separate the contribution of the edge from that of the screw dislocations. One of the reasons was that there is comparatively little dilation indicated around a screw dislocation when one uses isotropic elastic constants as the calculational basis. Consequently a smaller binding energy than for an edge dislocation is envisioned. However, a threefold symmetric dilatational field which surrounds a <111>-type screw dislocation results from using anisotropic elastic moduli [9]. Since the strain field is smaller than for an edge dislocation, and hence the density of conduction electrons is higher than it would be for an edge, the shift of lifetime from the bulk (trap free) value of 114 psec would be smaller, i.e., 142 rather than the edge value, namely 165 psec.

A single crystal was bent at room temperature in a die used to yield reproducible strains. A difference in the number of traps on the concave and convex sides are shown in Fig. 2. Aside from 114 psec, the value of 165 psec was found (single trap fit) for these bent specimens and associated with edge dislocations. The only previously reported value for a "dislocation" as 165 psec by Vehanen and his colleagues [10]. A further review of the literature yielded a number of instances with two separated lifetimes; one group within

INTRODUCTION

It has been stated in the literature [1-4] that a positron is not likely to be trapped in a dislocation because of the presumed low binding energy (ca.0.1 eV) and is more likely to annihilate in one of the jogs along the (edge) dislocation. As far as the authors are aware, there is no quantitative prior direct measurement which supports this contention. The results reported here on dislocation densities in Fe deformed by different means and measured by several techniques addresses this proposition. Some assumptions needed to invoke trapping by jogs or kinks are critically reviewed after it is noted that the number of measured positron traps is in good agreement with the number of dislocations determined independently on the same samples. The idea that rapid "pipe" diffusion of a positron occurs along the core of the dislocation is also discussed.

RESULTS AND DISCUSSION

Positron lifetimes have now been identified with annihilation in edge (165 ± 2 psec) and screw (142 ± 5 psec) dislocations in iron [5]. The fraction of the positrons annihilating in the shorter-lifetime trap, as well as in the longer, has been shown to increase monotonically with the deformation. One of the few studies on dislocations in iron in which an effort was made to study the dislocation density produced by cold-rolling is the classic work by Keh [6]. The density of mixed dislocations from that work is presented in Table I as a function of the reduction in area, along with the number deduced from the Doppler broadening runs of positron annihilation. The agreement is surprisingly good.

At the dislocation densities involved, an effective independent method of determination is the etch pit method. By using the etchant recommended by

POSITRON ANNIHILATION METHOD FOR DETERMINING DISLOCATION DENSITIES IN DEFORMED SINGLE CRYSTALS OF IRON

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ABSTRACT

Several crystals of high-purity iron were deformed to produce dislocation arrays, and the density of dislocations was estimated from changes in the lifetime spectra of the positrons. Interpretation of the spectra is made in terms of trapping and annihilation of positrons in both edge and screw dislocations with the densities of both being determined. The specific trapping rates were determined from concomitant Doppler-broadening and lifetime measurements on cold-rolled specimens. Tensile straining at 200 K produced more screw than edge components. The same specimens were etched and the density of etch pits was determined with the results/in good agreement with results of different independent determinations. Room-temperature cold-rolled single crystals were also studied by positron annihilation, and the density of dislocations was determined by TEM. These results are in good agreement with the study of dislocations in cold-rolled iron determined by Keh.

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20. Abstract (contd.)

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Bent Crystals$_1$
Single Crystals of Iron$_2$
Positron Lifetimes$_2$
Screw Dislocations$_2$
Edge Dislocations$_2$
Etch Pits$_2$
TEM Examination$_2$
Positron Annihilation$_3$
Dislocation Density$_3$

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FIGURE CAPTIONS

Figure 1. Comparison of the dislocation density in an iron single crystal which was deformed in tension at 200 K. Solid circles represent the total dislocation density as measured by positron annihilation and the open circles are the values from the etch pit measurements. Triangles and squares represent the number of edge and screw components measured with positron annihilation.

Figure 2. Doppler broadening lineshape parameter measurements for bent iron single crystals. In part (a) the peak-to-wing parameter P/W is plotted as a function of the curvature (i.e., the reciprocal of the diameter of the mandrel). In part (b) the same values of P/W are plotted versus the square root of the reciprocal of the diameter.

Figure 3. Transmission electron microscope photograph of dislocations on the (120) plane in a single crystal deformed in tension at 200 K. Possible Burgers vectors that would be associated with screw dislocations are shown. Strain was 2.5 percent.

Figure 4. Transmission electron microscope photograph of dislocations on (011) plane in a similarly deformed crystal strain is 9 percent. Since g = [011] some screw dislocations with (1/2)a[111] and (1/2)a[111] Burgers vectors may not appear.

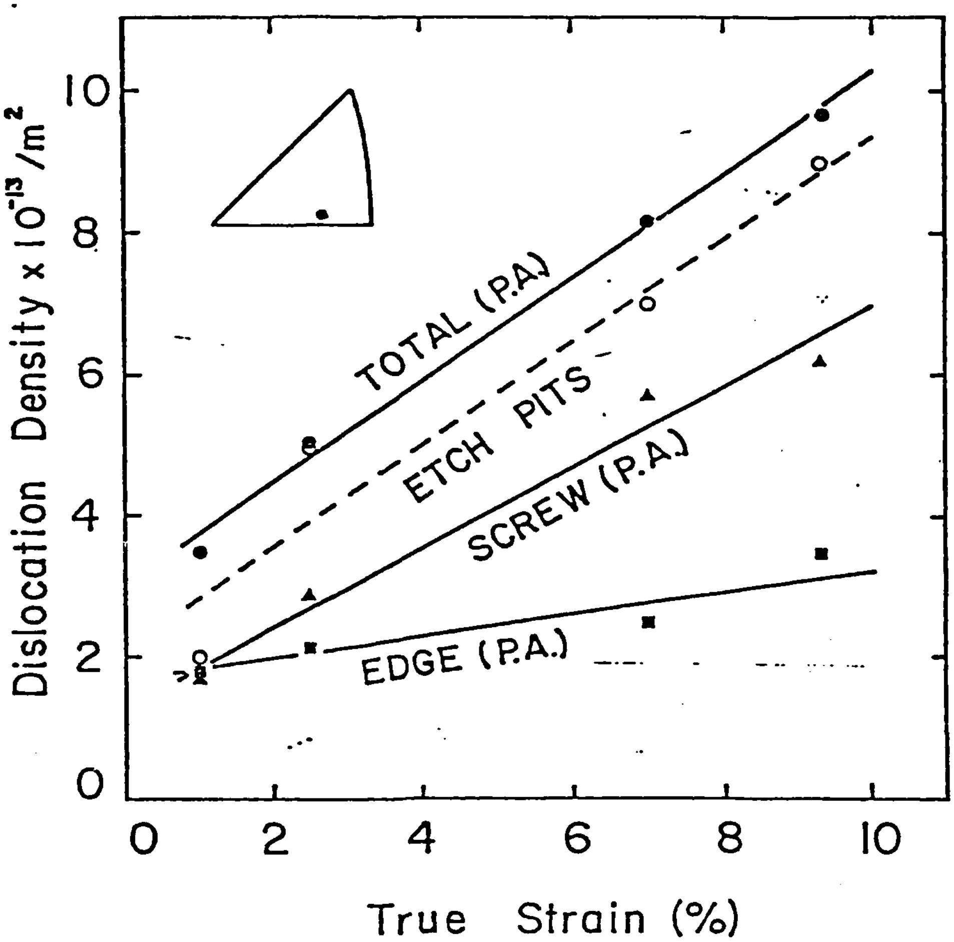
Figure 5. Comparison of the total (mixed) dislocation density as a function of the reduction of thickness determined from positron annihilation lifetime changes with the values determined from transmission electron microscope photographs of polycrystalline samples by Keh [6]. The TEM determination of the dislocation density using the present high-purity single crystals is also shown.

Cold-Rolled Iron

Reduction in Thickness (%)	$\rho_1 (m^{-2})$ $(\times 10^{13})$	$\rho_2 (m^{-2})$ $(\times 10^{13})$
5	5.8	5.95
10	9.2	9.27

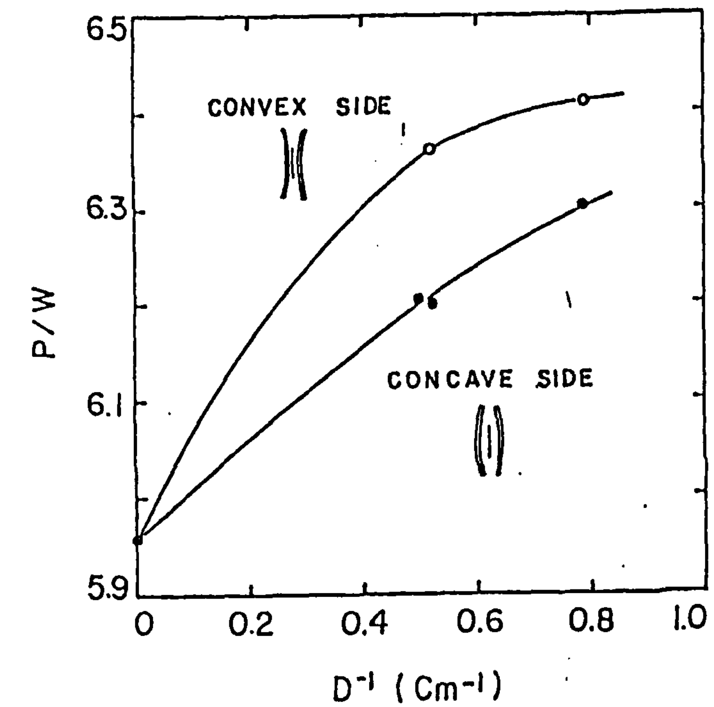
ρ_1: From A. S. Keh (1962)

ρ_2: Calculated from Doppler-broadening line-shape parameter(P) using $\mu_T = 1.1 \times 10^{-4} m^2/sec$ (Personal communication with A. Vehanen)

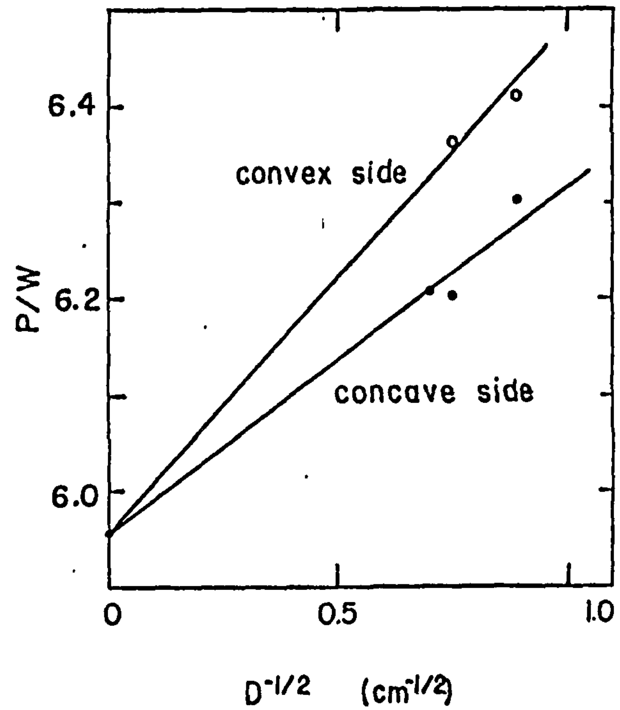


Dislocation Density x 10^{-13} /m^2
TOTAL (P.A.)
ETCH PITS
SCREW (P.A.)
EDGE (P.A.)
0
2
4
6
8
10
True Strain (%)

Yong-Ki Park
Fig 2



(a)



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Positron Traps 2	Hydrogen Embrittlement 3
Hydrogen Traps 2	Hydrogen Permeation 3
Hydrogen Occupation of Traps 2	
Dislocation Trapping 2	Iron Single Crystal 2

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The positron annihilation measurement was carried out with the pure iron single crystals deformed in various ways before and after hydrogen permeation. The positron trapping intensity was reduced more in the screw dislocation than in the edge dislocation by hydrogen charging. The trap occupancy by hydrogen was very close to the fraction of the reduction in positron trapping intensity.

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REDUCTION OF THE TRAPPING OF POSITRONS IN DISLOCATED SINGLE CRYSTALS OF IRON WHEN CHARGED WITH HYDROGEN

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The positron annihilation measurement was carried out with the pure iron single crystals deformed in various ways before and after hydrogen permeation. The positron trapping intensity was reduced more in the screw dislocation than in the edge dislocation by hydrogen charging. The trap occupancy by hydrogen was very close to the fraction of the reduction in positron trapping intensity.

INTRODUCTION

It has been known that a positron is trapped by dislocations in the deformed metals.[1,2] Recently it was found that the number of edge and screw dislocations can be determined separately by measuring the positron lifetime spectrum in the deformed iron single crystals.[3]

One of the applications of this non-dêstructive microstructure characterizing method is the study of hydrogen trapping phenomena in metals. This study is important to understand and control the hydrogen embrittlement in metals.

Hydrogen permeation method is one of the most frequently used techniques in the study of hydrogen trapping. The major diffuculty in this method is that there is no conclusive identification of trapping site.

Positron annihilation method is one of the complementary techniques to solve the above difficulty in the hydrogen permeation method. Hydrogen in metals is known as a screened proton[4]. Since it has the same charge as the positron, a hydrogen which is trapped in the defect will repel a positron and will reduce the probability of a positron trapping in the same type of defects.

In the present research, low concentration of hydrogen was charged in the pure iron single crystals. The positron lifetime and Doppler broadening was measured in the hydrogen-charged specimens in order to identify the trap site and determine the trap density.

EXPERIMENT

Pure iron single-crystal specimens were prepared and purified by circulating ZrH_2 purification system. They were deformed by various ways such as bending, cold rolling at room temperature, and elongating at 200 K.

Hydrogen permeation measurement was carried out using the Devanathan and Stachurski type electrochemical cell.[5] Experimental details are decribed in elsewhere.[6]

Hydrogen - charged specimens were quenched in the liquid nitrogen. Positron annihilation lifetime and Doppler broadening line-shape were measured concomitantly with the specimens before and after hydrogen charging. In order to keep the hydrogen in the specimen, the positron annihilation measurement was carried out with the specimens in the liquid nitrogen Dewar.

RESULTS AND DISCUSSION

Figure 1 shows the total positron trapping intensity of trap site in hydrogen-charged and uncharged iron crystals which were deformed in tension at 200 K. The total positron trapping intensity was decreased about 20% by small amounts of hydrogen (ca. 0.3 atomic ppm).

It was found that the screw and edge dislocations could be determined

separately by the measurement of positron lifetime.[5] When hydrogen was charged in low temperature deformed iron the positron trapping rate decreased both in screw and edge dislocations. Figure 2 shows the positron trapping rate in screw and edge dislocations before and after hydrogen charging.

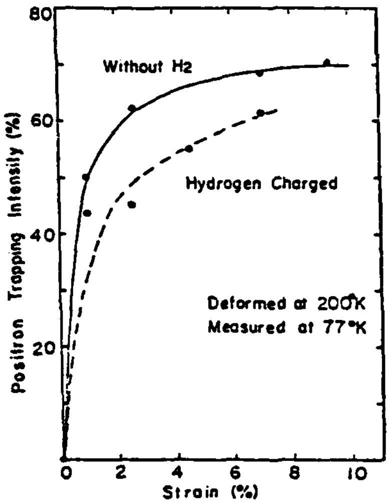

Figure 1. Total positron trapping intensity at trap site in hydrogen charged and uncharged iron single crystals which were deformed in tension at 200 K.

The hydrogen effect was larger in a screw dislocation than in a edge dislocation. This is probably because the binding of positrons is weaker in a screw dislocation than in an edge dislocation since the dilatatonal field around a screw dislocation is smaller than that around an edge dislocation.[7] Hence, the modification of the local strain field and the electron density profile by the trapped hydrogen could be more effective in reducing the positron trapping in the screw dislocation than in the edge dislocation.

The hydrogen trap density was determined from the hydrogen permeation experiments using several different analyzing models.[8-10] This results were compared with the dislocation density determined by positron annihilation and by transmission electron microscopy.

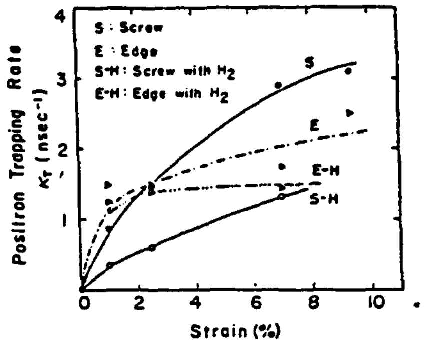

Figure 2. Positron trapping rate at screw and edge dislocations in hydrogen-charged and uncharged iron single crystals which were deformed in tension at 200 K.

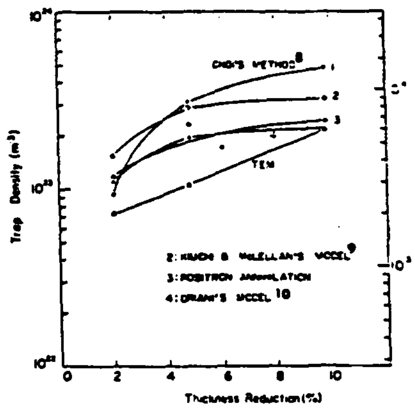

Figure 3. Hydrogen trap density in room-temperature cold-rolled iron single crystals.

The dislocation density and hydrogen trap density was correlated by assuming that there is one trapping site per unit cell length along the dislocation line.[9] The agreement between three independent measurement was good as shown in Figure 3.

The reduction of the positron trapping intensity of the trap in the hydrogen-charged iron could be anticipated because when a hydrogen atom which has the same charge as a positron occupies the trapping site it will repel a positron.

Table 1. Comparison of the trap occupancy by hydrogen determined by hydrogen permeation method and positron annihilation technique in iron single crystals deformed in tension at 200 K.

True strain (%)	Trap degsity N_T, m^{-3}	Average Hydrogen concentration C_{av}, H_{atoms}/m^3	C_{av}/N_T	$\Delta I = (I_D - I_{DH})/I_D$
1	8.5×10^{22}	1.44×10^{22}	0.17	0.14
2.5	1.64×10^{23}	2.48×10^{22}	0.15	0.27
4.4	2.78×10^{23}	2.66×10^{22}	0.10	0.14
6.9	4.14×10^{23}	1.89×10^{22}	0.05	0.11

*I_D: positron trapping intensity in deformed specimen (without hydrogen)

I_{DH}: positron trapping intensity in deformed and hydrogen-charged specimen

In Table 1 the quantitative comparison between the trap occupancy by hydrogen atoms and the reduction of the positron trapping intensity was made. In this table the trap occupancy by hydrogen was defined by the ratio between the average hydrogen concentration and hydrogen trap density which were determined from hydrogen permeation experiment. The equivalent parameter in terms of positron trapping intensity can be defined by

$$\Delta I = (I_D - I_{DH})/I_D$$

where I_D and I_{DH} is the positron trapping intensity in a specimen deformed but not charged and that in a specimen deformed and charged, respectively. This parameter depends on the hydrogen occupancy in the defect and also on the intensity of repulsion of positron by a trapped hydrogen atom.

According to the theoretical calculation by Jena *et al.*[11] the electron charge pileup around the proton is localized. Therefore it is anticipated that hydrogen trapped in the defect will reduce the positron trapping intensity by about the same fraction as the hydrogen occupancy.

As seen in Table 1, hydrogen occupancy determined by hydrogen permeation method is actually very close to the ΔI as anticipated above. Further, the hydrogen occupancy along the dislocation obtained in the present research is also in good agreement with the reported value of Gibala[12], namely about 0.25. This value was obtained from his internal — friction measurements.

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Lifetime and Doppler-broadening measurements for positron annihilation in substoichiometric nickelous oxide have been made concomitantly from liquid-helium to room temperature. The concentration of cation vacancies is readily controlled by altering the ambient oxygen pressure while annealing the crystals at 1673 K. It was found that neither of the three lifetimes observed or their relative intensities varied significantly with the oxygen pressure, and the bulk rate only increased slightly when the specimen was cooled from room to liquid-helium temperatures. These results are interpreted as indicating that some of the positrons are trapped by the existing cation vacancies and a smaller fraction by vacancy clusters.

INTRODUCTION

In recent years, there have been a number of measurements of positron annihilation for various nonmetallic crystals. Complicated lifetime spectra have been observed for ionic solids and the effects of lattice defects deduced.[1-3] The shortest component τ_1 ranges from 132 psec for LiBr to 347 psec for NaI and is usually associated with annihilation in the bulk of the crystal. Dupasquier[4] recently reviewed the results for a series of alkali halides and other ionic crystals such as oxides; it is not clear that the bulk lifetime is always the shortest or the most-intense component that is present. A second lifetime component which increases with the average number of anions per unit volume was observed to range from 297 psec for LiF to 776 for NaI. There is an intermediate lifetime in the range of 136 to 183 for the four compounds, CsBr, KI, RbI and CsI. The cause for the several lifetimes and decay modes is not established unequivocally.[4] There is a tendency among workers to ignore any third component which may be 1 or more nanoseconds long, even though there may be good statistical grounds for its being significant.

In analogy with metals, the second longest lifetime component is associated with the trapping of the positron by a lattice defect. It is definitely associated with a region where the electronic charge density is reduced. The most likely candidate would appear to be a cation vacancy. Studies on intentionally deformed crystals suggest that positrons may also be trapped by dislocations.

In the literature on lattice defects, cation vacancies are described as being singly or doubly ionized. In the singly ionized state, an electron is localized near the vacancy and an electron hole is also in the vicinity. The latter could be an increased valence (from 2 to 3) of nearby cations or it could be a reduction in the charge on the O^{-2} ion in NiO. Theoretical calculations indicate that an oxygen ion could readily lose an electron from the closed shell and have it delocalized towards the vacant lattice site, i.e., a spreading of the charge distribution towards the low-density region. Such details are seldom discussed, since the approach taken very successfully by Wagner[5] is to treat lattice defects as dilute solutions with the host lattice acting as the "solvent" and apply the Law of Mass Action.[6] Based on chemical intuition, it is assumed that the anions have a fixed valence whereas the cations have variable ionic charges.

Because of the redistribution of the electrons near the vacancy, it is not clear whether one or two traps (one much weaker than the other) might be expected in an oxide such as NiO. Our experiments were further prompted by the fact that the formation of cation vacancies in NiO has been studied many times in the last 20 years[7-8] and various authors agree about the effect of vacancies on electrical conductivity and related physical properties and that the vacancy is singly ionized.[9]

EXPERIMENTAL DETAILS

A slice of a NiO boule was obtained from Argonne National Laboratory; such boules have been used in tracer diffusion measurements and the total impurity content was of the order of 10 ppm. After cleaving, the mating surfaces were oriented and ground so that the normal was within a few

degrees of [100]. The two pieces were annealed for several days in air at 1691 K. The estimated vacancy concentration was 7×10^{-4}. In the later experiments, annealing was done at 1670 K for 100 h in high-purity N_2 gas which intentionally contained 7.5 ppm of O_2. This fugacity of oxygen would correspond to a concentration of 0.5×10^{-4} singly ionized cation vacancies.[9]

The positron source was deposited by drying a drop of ^{22}NaCl solution on a Kapton film. This polymeric material has been found to have a single temperature-independent lifetime of 382 psec.[10]

For the low-temperature runs, the source holder was inserted between the two portions of the crystal and they were secured to a copper block which was in intimate contact with the central column of an evacuated Dewar. The photomultipliers, the Ge(Li) detector, and electronic circuits are those routinely used at Brookhaven National Laboratory.

RESULTS

Values for the short lifetime component are plotted in Fig. 1 and the nitrogen (low oxygen partial pressure) anneals. No large changes in τ_1 are evident. The question of whether the data were fitted better with two or three lifetimes is examined in Table 1. The values of χ^2 are improved by including a long lifetime. The remainder of the values in this report are based on this third component τ_3 being included in the analysis. Typical values are presented in Table 2. The temperture dependence of τ_3 and the intensity I_3 are illustrated in Fig. 2. Only about 20% of the decays come from lifetimes in the vicinity of 350 psec, and this lifetime value was insensitive to differing atmosphere or temperature. It was difficult to extract an accurate value from the data because of the proximity of the lifetime associated with Kapton, namely 382 psec. The value of the second component reported by Bertolancini et al.[11] and by Chiba et al.[12] namely 353 and 350 psec, respectively, is frequently associated with annihilation in lattice defects.

Concomitantly, the change in the shape of the gamma ray peak was studied. The changes observed in the S parameters for the differing cation concentrations or measurement temperatures were too small to be of interest.

DISCUSSION

The present measurement of τ_1 for annihilation in the bulk is consistent with the value obtained by Kunz and Waber[13] who calculated the positron and electron bands in NiO using a Restricted Hartree-Fock formalism. The annihilation rate is proportional to the overlap of the positron and electron charge densities. By using the band orbital for three high-symmetry points in the Brillouin Zone of NiO, the lifetime was calculated to be 150 psec, (for the lowest-energy point, (Γ_1), 170 and 160 psec. It is probable that only the point at the center of the Brilloium Zone is occupied. The polaron model was used to estimate correlation effects; the annihilation rate was increased by a multiplicative factor (1 + a). The constant is that used in connection with the band electronic structure of numerous alkali halides and ionic crystals. This theortical value of τ_1, namely 150 psec, agrees very well with the experimental values reported herein. However, other models for dealing with correlation have been presented,[14-15] and they are more nearly first-principles calculations.

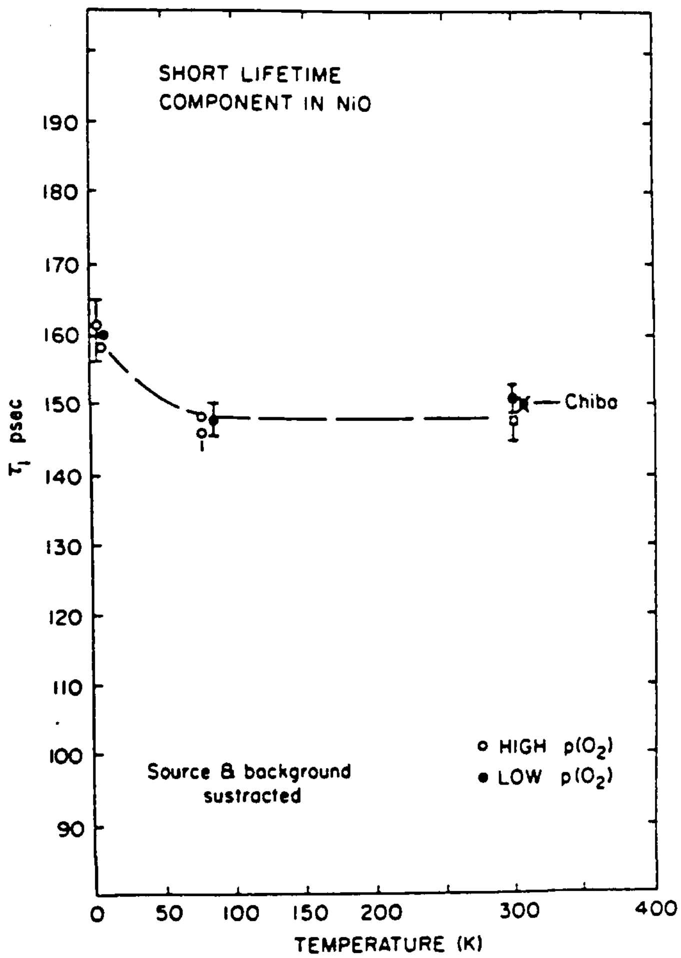

Fig. 1: Temperature dependence of bulk lifetime of annealed NiO.

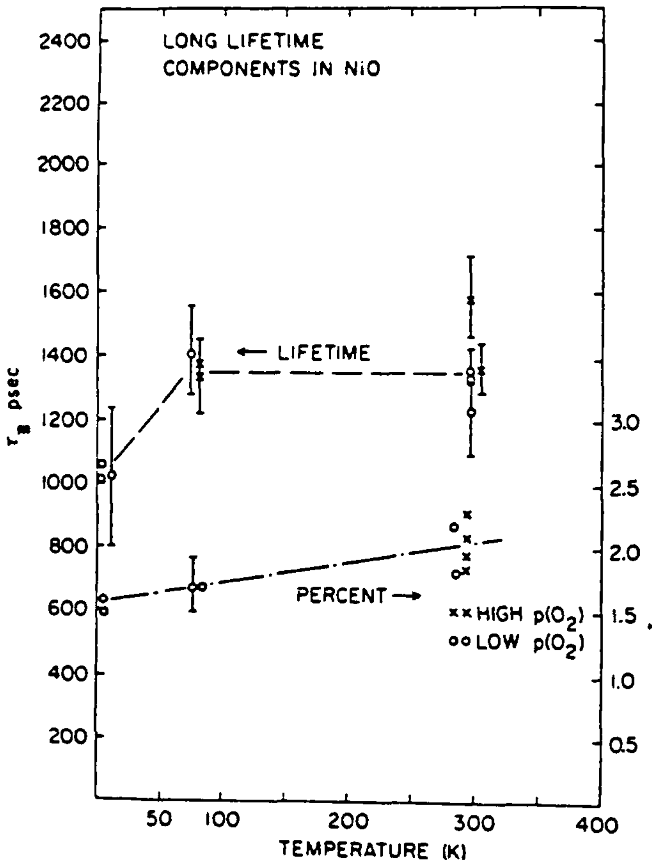

Fig. 2: Temperature dependence of longest lifetime and intensity I_3.

Table 1. Comparison of Fitting Positron Decay Data with Two and Three Lifetimes : Values of τ_1

Run	Temp.	Two Lifetimes χ^2	τ_1	I_1	Three Lifetimes χ^2	τ_1	I_1
21	Room	4.65	112±6	84	1.54	136±3	86
23	Room	2.38	129±5	84	0.89	140±2	85
37	Liq. N_2	2.24	117±6	72	0.97	154±4	77
41	Liq. N_2	2.99	115±6	72	1.02	154±4	78
01	Room	2.50	113±8	72	1.05	153±7	77
03	Room	4.66	111±7	72	1.34	153±4	77
01	Room	3.64	106±8	74	1.04	148±2	78

Kunz and Waber[16] used a computational procedure, similar to that used by Surrat and Kunz[17] to study the adsorption of H on NiO, to investigate the trapping of a positron in the vacancy of NiO. They found an eigenvalue of 8.1 eV and calculated a lifetime of 280 psec.

The reported temperature dependence of annihilation,[18] in either NaCl or KCl is also small. The τ_1 of NaCl only increases by about 13% when it is heated from 290 to 973°C and the other spectral components (lifetimes and intensities) are virtually unchanged. Cooling KCl from 290 to 77 K only increased τ_1 by about 10%. The present results for NiO agree with this trend.

One explanation of the third component is that some of the positrons are being annihilated within voids. It is not uncommon to find vacancy clusters or voids in single crystals of NiO by Transmission Electron Microscopy as Sears[19] and Dubois[20] reported.

In view of the large number of cation vacancies present in NiO and the large binding energy, it would seem reasonable to conclude that all of the positrons are trapped and they would not be expected to become thermally detrapped at room temperature. Our results of 80% annihilations with a lifetime of 150 psec associated with the bulk seems to refute this.

CONCLUSION

The shortest measured component of the lifetime of positrons in NiO, namely 150 psec, is in close agreement with the value calculated by Kunz and Waber for annihilation in the bulk to which we attribute the majority of the annihilations. The second component, near 350 psec, was difficult to determine due to the proximity of the lifetime of Kapton used in the positron source, but is similar to the theoretical value of 280 psec. It was insensitive apparently to changes in vacancy concentration.

ACKNOWLEDGMENTS

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In Fig. 10, the slope of the screw component is similar to that of the total dislocation density but the edge component has a much smaller slope which implies that the screw dislocation is dominant during deformation at this temperature.

2. Concerning the R Parameter

Taking note of how different electron densities are involved in annihilation of a positron when in a specific defect trap, Mantl and Triftshauser[52] developed an R parameter which was characteristic of the defect. An important advantage is that it is essentially independent of the concentration of the defect involved. This parameter can be written as

$$R = \frac{| P_T - P_F |}{| W_T - W_F |} \qquad (4)$$

While it is specific to a given type of defect one finds it difficult to establish what defect is present by comparing results with published R values since most authors do not specify the energy ranges they used for P and W in analyzing their Doppler data. In Figure 12, the R parameter is shown for a series of single crystal specimens deformed in tension at 200 K. As the strain increases, the value of R approaches 1.8. The specimen from crystal 2 (shown as a a filled square) which was held at 300 K for several hours after deformation did not yield a value significantly different from the other values. These measurements were made at 77 K.

Specimens deformed at room temperature are the subject of the next figure. The R values for specimens bent at room temperature are plotted in Fig. 13 as a function of the inverse of the diameter of the mandrel. These values are somewhat smaller than in the experiment just cited. One possible reason is that they were obtained in runs at 300 K. The measurements made at 77 K are plotted as squares in this figure and their R values lie above 1.8. However, it is generally found that R is insensitive to temperature in this range. The difference may reflect the statistical variability of the experiment rather than any inherent cause.

DISCUSSION

Several subjects are taken up under this heading. First, the question of dislocations versus jogs which is central to this investigation. Next, the resolution of two different lifetimes for a dislocation is discussed. Then the specific trapping rate the authors obtained is compared with values which were previously reported but not emphasized. Then some results obtained by others are summarized. General suggestions for further

measured in a variety of techniques such as electron microscopy, x-ray line broadening, and hydrogen permeation. Their results were internally consistent. Since it is difficult to locate this journal, one of their figures is presented as Fig. 11. They also separated the densities of edge and screw dislocations based on the profile of certain x-ray diffraction lines.

In the following calculation, the total dislocation density was obtained from their values for electron microscopy at a same two values of strain. In addition, the ratio of screw to edge dislocations was obtained from their x-ray results. By using the total dislocation density for 5 and 10 percent from Yamakawa *et al.* with values of κ_T determined in our lifetime spectra, the specific trapping rate for a generic or "average" dislocation was calculated. This is a reasonable assumption to make initially in the absence of any prior determination. The total density was then calculated from the P parameter data of the Doppler-broadened lines from the same two specimens using the "average" μ_T just obtained. The numbers of edge and screw dislocations were estimated using the ratio they gave. Thus two trapping rates were separated for the edge and screw components. These results are presented in Table III and discussed below. In this way the data were normalized.

As was anticipated the specific trapping rate we have obtained for a screw dislocation μ_S is smaller than that for an edge dislocation μ_E. Kuramoto *et al.*[49] also briefly discussed that such a result would be reasonable to expect. For cold-rolled specimens the agreement is good for the values of the total observed by Yamakawa (in Column 2) and those calculated from the Doppler line-shape data (rather than the lifetime measurements) which are presented in Column 4. These specific trapping rates were used with their κ_T values to estimate the dislocation density of iron single crystals produced in two ways, both in bent specimens and in those deformed at low temperature. The specific trapping rate for an edge dislocation was used in calculating the dislocation density in bent specimens as shown in Table IV, the dislocation density on the convex side (tension side) is somewhat higher than on the concave side (compression) side.

In addition, the results for low temperature specimens are plotted in Fig.10. The dislocation density increases linearly as a function of the true strain. It was reported[50] that in the early stage of deformation there is a linear portion in the relationship between the dislocation density and the strain. This linearity was satisfied up to at least a true strain of 10% [50,51] The total dislocation density measured by the etch-pitting technique is somewhat lower than that measured by positron annihilation. However, the slopes of the density vs strain curves are quite similar to each other. In interpreting the etch-pit technique some of the dislocations might not be revealed because a short etching time was used to increase the resolution, and therefore the actual dislocation density could be higher than the number of etch pits. When this is considered, the total dislocation density measured by positron annihlation is in very good agreement with the density of etch pits.

the whisker. Since these were undoubtedly due to screw dislocations and only the 142 psec type trap was found in the deformed whiskers, this lends credence to the assignment of 142 psec to the screw dislocation.

In view of this finding, low-temperature-deformed single-crystal sheet specimens were examined. The same specimens from Crystal 1, which had been measured by positron annihilation, were chemically polished in a solution of 80% hydrogen peroxide (of 30 % aqueous solution), 5 % hydrofluoric acid (48% solution) and the balance water. Then they were etched in the potassium sulfate + sulfuric acid mixture recommended by Shemenski, Beck and Fontana[44]. By accident, one optical microscope picture contained a sub-grain boundary. It appeared that there were etch pits on one side of the boundary and not on the other. Actually this appearance was due to the fact that the individual pits could not be resolved by an optical microscope on the side thought to be "free" of etch pits. This could be discerned by examining a replica in the transmission electron microscope.

In order to resolve the individual etch pits so they could be counted, the etching time was reduced to 20 seconds. A two stage carbon replica was made of the etched surface and the former shadowed. The increase in the number density of pits as photographed in a transmission electron microscope can be seen in Figure 9. In counting the etch pits, enlargements of the the original photographs were used. Pictures were prepared from several different areas of the specimens and the average number of pits was divided by the area in the photograph. This number was taken as the total dislocation density. Results are presented in Table II and he data are plotted in Figure 10 to show the linear dependence on the true strain.

ANALYSIS OF THE DATA

This section consists primarily of the determination of the specific trapping rates after the number of dislocation of the edge and screw types had been determined.

1. Determination of Dislocation Density

In Equations (2) and (3) above, a trap concentration can be calculated if the specific trapping rate μ_T for a specific kind of a trap is known. Unfortunately there are no published values of the specific trapping rates for dislocations in iron. The present concomittant measurements of both the Doppler profile and the lifetime on the same specimen offer the possibility of using the two equations to determine two unknowns. Note that κ_T in Equation (2) can be determined from positron lifetime spectra and F can be replaced by the P parameter determined from the Doppler broadened lineshape to yield a second value.

The authors have estimated the specific trapping rate from cold-rolled specimens in the following manner. Yamakawa *et al.* reported the dislocation density in pure iron[47] and in mild steel[48]: specimens which were deformed at room temperature and

perature. Crystal 2 after straining 4.5% had a ratio of two trap intensities of 4.5. It was permitted to warm up and was kept for 48 hours at 300 K. The ratio of I(screw) to I(edge) decreased to 1.5. This is certainly consistent with the anticipated disappearance of screw components.

4. Positron Annihilation in Iron Whiskers

A number of whiskers of iron were prepared by decomposition of ferric chloride hydrate in gas stream as described by Wayman[42]. A number of these whiskers were studied using positron annihilation and scanning electron microscopy after using an etchng solution to reveal etch pits, In the as-grown condition they were found to contain a small but measurable concentration of edge and screw dislocations rather than the single axial screw dislocation which is generally expected. This is consistent with the preponderance of x-ray and electron microscope studies of iron whiskers[43-45]. Difficulties were encountered in resolving a single or "mean" lifetime of 120.1 $\pm$ 3.2 psec which gave a X^2/ν value of 0.97 when all of the fitting parameters were "floated." After fixing 114 psec for the bulk, a single trap gave 157 psec with an equivalent X^2/ν value. Subsequently this was resolved into two components with the intensity fractions for 142 psec was 12 % and for 165 was 21 % As will be seen shortly, these data correspond to approximately 6.8 and 12.1 x 10^{12} dislocations per square meter.

Subsequently, a number of whiskers were deformed in torsion i.e., were twisted roughly 720°. A set of new samples was prepared from these. In the early stages, a very significant tail was observed with a lifetime approaching 2000 psec. It was suggested by Nielsen[34] that many of the positrons were not striking the whiskers. By improved packing of the twisted whiskers and making sure that plastic tape was not exposed, this tail was eliminated. The measured single lifetime was 142 psec and the percentage of positrons annihilating (in the single 142 psec trap) was 62%.

At this stage, we do not see contrary evidence to our assignment of 142 $\pm$ 5 for screw and 165 $\pm$ 2 psec for edge dislocations. However, most writers have indicated that positrons probably annihilate primarily at jogs along edge dislocations. A similar possibility of annihilating at kinks on screw dislocations although not mentioned is an obvious extension. Independent evidence for, at least, the total number of dislocations was sought and sis discussed next.

5.Development of Etch Pits and their Examination

Etch pits were also formed in the torsionally deformed whiskers using either the Nital and the sulfate solution recommended by Shemenski, Beck, and Fontana[44]. The etched whiskers were examined in a scanning electron microscope. A linear array of hexagonal-shaped pits of the <111> type were observed which had their dislocation cores roughly perpendicular to the long axis of

left-hand panel is the peak-to-wing parameter P/W from the Doppler-broadening measurements. The fraction of positrons annihilating in the "trap" I_2 is shown in the right-hand-side as Fig. 4b. The lifetime in this trap was 155 ± 3 psec. In contrast to the lifetime found for the cold-rolled specimens, the single lifetime in traps of the bent specimens was 165 ± 5 psec. Reasonably straight lines are obtained when these positron data are plotted versus the square root of the plastic strain ϵ. Typical results are shown in Figure 5 where the peak parameter P from the Doppler broadening was used

3. Effect of Tensile Deformation at Dry-Ice Temperature

There is general agreement[37,38] that screw dislocations are produced primarily in high-purity iron samples which are deformed at 200 K or lower. To investigate the possibility that the annihilation rate in screw dislocations might be different from that of an edge, the following experiments were done.

Stress - strain curves for two different orientations are shown in Fig. 6. There is some evidence of a weak yield point. The insert shows the orientation of the single-crystal sheets. The orientation of zone-refined single iron crystals used by Kimura and Kimura [39] (whose work will be discussed below) is indicated by point K in Figure 6. When some of the lifetimes from these specimens were analyzed, the value of approximately 142 psec (for a single-trap fit) emerged. It is important to note that the fraction of positrons annihilating in this trap increased monotonically with the tensile strain ϵ.

The fraction of positrons annihilating at 77 K in the traps created by deformation is plotted (for a single-trap fit) versus strain ϵ in Figure 7. Then using a two-trap model, two curves of κ_T are presented in Figure 8; one for 142 and the other for 165 psec components. As noted above, the relative trapping rates κ_T are directly proportional to each dislocation density ρ_T. As has been reported by Kubin[40] the number of edge components rises initially more rapidly than the number of screw components, and as observed here, after about 2 or 3% elongation, this situation reverses and further deformation of iron at 200 K produces mainly screw dislocations. In the absence of any definitive information about the specific trapping rate from dislocations, the authors used an "average" value of the specific trapping rate μ_t to estimate total density of dislocations. Subsequently the edge and screw dislocations were handled separately.

The smaller dilatational field *vide infra* around the core of the screw dislocation compared to that of an edge dislocation[41], would suggest that μ_T would probably be smaller for the screw components. Consequently the number of screw dislocations would be relatively even larger. There appears to be general agreement among electron microscopists that the collection of screw dislocations formed at 200 K in iron becomes motionally (mechanically) unstable when the temperature approaches room tem-

of dislocations measured by x-ray line breadth. If a specific value exists for both the peak and the wing for a given type of trap, then a $(P/W)_j$ exists for the j^{th} type of trap and one can form a linear combination of these values. Nevertheless, the P parameter was used in a number of cases below, in response to these objections and in an effort for conformity and simplicity, rather than solely for reliability.

The trap concentration C_T can be expressed[29] by another equation

$$C_T = \frac{\lambda_T (F - F_B)}{\mu_T (F_T - F)} \qquad (3)$$

where F is any characteristic property or parameter of the positron annihilation process which is a linear function of the positron state. Then F_T and F_B are F values of the trap and trap-free bulk, respectively.

The line shape was found to be dependent on the counting rate as was observed recently by Nielsen.[34] As the counting rate decreased, the value of P increased. However, these changes become very small when the total counting rate was near 2×10^3 sec^{-1}. This counting rate was adopted.

RESULTS

A variety of results are reported in this section such as the lifetimes of the two basic types of dislocations, the annihilation characteristics of iron whiskers, as well as the results of etching the specimens to develop etch pits and counting them by means of replicas examined in a transmission electron microscope.

1. Cold-Bent Specimens

Single-crystal specimens were bent to a fixed radius by means of a machined cylinder mating a fixed mandrel. The concave and convex sides of the bent specimens were studied separately. In Figure 3a, typical results are plotted versus D^{-1}, and in 3b versus the square root of D^{-1} where D is the diameter of curvature of the mandrel used, which in turn is linearly dependent on the strain ϵ. Because of the formation of redundant dislocations during the deformation, the number on either side should not be expected to be as small as the minimum number of dislocations needed to produce the curvature. The latter depends on the net number of dislocations. The densities determined for the two sides of a specimen are presented in a later section.

2. Cold Rolled Specimens

Some of the early results for cold-rolled single-crystal specimens are presented in Figure 4. The Y axis of the

In order to see the effect of total counts, the lifetimes were measured with well-annealed iron specimens using progressively longer counting periods to acquire more total counts. It is interesting that the lifetime does not change significantly even at low total counts (such as one half million). However, at higher counts the values of X^2/ν improve. Also the scatter of the data and the standard deviations become smaller. Therefore, more than one million counts were taken in almost all of the experiments.

The positron lifetimes obtained (assuming only one trap) for a repetition of runs with the same specimen differ only by 2 psec and results of measurements on different specimens vary within the same range. The mean of all the lifetime determinations for annihilation in the bulk is 114 ± 2 psec. This value is in good agreement, e.g., within experimental limits of the four independent values reported for iron, namely 117 psec reported by Doyama and Cotterill[3], 111 by Cao Chuan *et al*,[30], 110 psec by Vehanen and Hautojarvi[12,13], and 108 by Van Brabander *et al.*[31].

In addition to the good reproducibility of these measurements, the question of whether the numbers obtained from these measurements at Northwestern University are accurate can be favorably answered. Well-annealed, high-purity aluminum specimens were measured at Northwestern University and at Brookhaven National Lab. The two lifetimes were 164.1 ± 3.7 and 162.5 ± 1.6 psec respectively. Not only are these two values very close to each other, they are in excellent agreement with the reported value for Al of 163 psec by Schultz *et al.*[32], and of 161 ± 2 by Goland and Snead[33],

B. Doppler-Broadening Measuring System

The line shape of the Doppler-broadened γ rays from positron annihilation was measured with a high-purity germanium detector (Ortec Model GEM-10175). The energy resolution (expressed as FWHM) was 1,70 keV at an energy of 1.35 MeV from ^{60}Co.

The Doppler-broadened line-shape was analyzed using line shape parameters after subtracting the background expressed as a two-sided error function discussed by Jorch and Campbell.[34]. In simplified line-shape analysis, the Doppler spectrum is not fitted, but areas under the curve in limited energy ranges are compared. The peak-to-wing parameter P/W used here is the ratio of the area under the central 19 channels to the area of the two wing portions centered 38 channels on either side of the peak channel of the Doppler curve. The absolute value of P/W is larger than P and hence is more sensitive to changes in the defect concentration. It has been argued that since P/W can be represented as a ratio of two linear functions of the defect concentration, the simple quantity P is preferable. A small amount of algebraic manipulation will show that the ratio is, to first approximation, a linear function with a small remainder. Evidence will be presented below that Byrne and coworkers[35] found that P/W is a linear function of the density

In analyzing the spectrum, the right-hand-side slope of the resolution function was fixed to see if there was any effect of the shape of the resolution function on the lifetime and/or on the X^2/ν values: here ν is the number of degrees of freedom in the statistical analysis. Figure 2 shows the variation of the lifetime (actually the time difference) and X^2/ν as a function of the right hand slope of the resolution function (SR). The lifetime decreases as SR is increased because a larger fraction of the time spectrum is deconvoluted as a resolution function. The X^2/ν parameter has a minimum for SR values around 45 psec and increases rapidly if SR is increased further. If we choose the best X^2/ν condition (in this case the minimum value of X^2/ν) the time difference between the two γ rays' generation in ^{207}Bi is 185.2 ± 0.5 psec which is in quite good agreement with MacKenzie's value[27,28].

In this figure, one sees that the lower value of SR leads to a relatively small change (ca. 1 psec) in the lifetime and also in X^2/ν. A fitted value of SR less than 45 psec would, in general, be acceptable as causing a minimum uncertainty in the measured lifetime. Therefore in analyzing the lifetime spectra the initial value of the SR was chosen near 45 psec and the other parameters were initialized using the values obtained with ^{207}Bi. The resolution function obtained with the ^{207}Bi spectrum is similar to that for ^{22}Na, which is plotted in Figure 2.

(i.) Mathematical Treatment of the Lifetime Data

The intensity in the lifetime spectra of a trapping site (in which the positron lifetime is $1/\lambda_T$) is given by the expression

$$I_T = \frac{\kappa_T}{\lambda_* - \lambda_T - \sum_J \kappa_J} \qquad (1)$$

$$\text{where } \kappa_T = \mu_T C_T. \qquad (2)$$

Here κ_T is the positron trapping rate in the specific trap, $1/\lambda_T$ is the lifetime in the bulk, μ_T is the specific trapping rate for the trap, and the summation runs over the trapping rates of the other j traps.

Before fitting the spectrum to the trapping model, a source component and the background were subtracted. Source components in several annealed specimens were initially measured and an average value was used as a trial value for the deformed specimens. When not in the liquid nitrogen Dewar, the source component was 340 psec with an intensity of 5.5 %. When the annihilation spectrum was measured at 77 K in the Dewar, the source component increased to 570 psec and an intensity of 8 %. When the source component was not fixed during fitting, it varied only ± 10 psec.

(ii.) Reproducibility and Accuracy of the System

room temperature. Crystals #1 and #2 were taken from this rod.

2. Positron-Annihilation Measuring Equipment

A. Positron Measuring System

Positron lifetimes were measured by a fast-fast system. This system measures the time interval between (i) the arrival of the first γ ray of 1.28 MeV which is emitted from the ^{21}Ne nucleus about 10 psec after the positron is emitted from the ^{22}Na nucleus and (ii) the detection of one of the two 0.511 Mev γ rays which results from the annihilation of the electron-positron pair.

The positron source was prepared from an aqueous solution of $^{22}NaCl$ which was evaporated onto a thin titanium foil (ca.1.13 mg/cm^2, covered with a like foil and then sandwiched between two identical specimens. This sandwich was held in place by wrapping it in aluminum foil. Since the two γ rays are emitted at nearly the same time, the measured time spectrum may be a close approximation of the prompt resolution curve[21,22]. However, because the energy spectrum of ^{22}Na is different from that of ^{60}Co, there might be some deviation between the natural resolution and the measured prompt curve using ^{60}Co.[23,24]

Annealed, bent, and cold-rolled single-crystal specimens were measured at room temperature. Some of the above specimens as well as some which were deformed at low temperature were measured in a specially designed Dewar at liquid nitrogen temperature in order to retain their dislocation structure. For room-temperature measurements, a 10 µCi source was used. In order to obtain a greater counting rate , a stronger source of about 80 µCi was used in the Dewar. In essentially all of the experiments the positron lifetime and Doppler broadening were measured concomitantly

Figure 1 shows the time spectrum measured with ^{60}Co and the fitted function in well-annealed iron single crystals with ^{22}Na. The resolution function we used was a Gaussian with a double sided exponential[25,26]. The full width at half maximum (FWHM) of the ^{60}Co is 299 psec and that of the ^{22}Na is 301 psec.

In order to test the timing system further, the lifetime of ^{207}Bi was measured. This isotope is known to be an excellent standard, since it emits two γ rays of 1.06 and 0.57 MeV which are similar to the γ rays of ^{22}Na and the lifetime of the metastable state is 187 psec.[27,28]. Moreover, there is no source component when using ^{207}Bi which might cause error in the positron lifetime measurement. A 21.4-µCi ^{207}Bi source was placed between two scintillators and the time spectrum was collected without changing the settings of the lifetime system.

between the dislocations and the positron traps is verified. These three topics will be discussed in greater detail in a later paper.

In this paper we will direct our attention to the effectiveness of trapping by edge and screw dislocations, and the ability to determine the number of dislocations of each type per unit area. A discussion of the effect of other defects such as monovacancies will be deferred.

EXPERIMENTAL PROCEDURE

In this section, the preparation of the specimens and their orientation as well as details of the measuring techniques and establishing the measurement precision will be presented.

1. Preparation of Specimens

The single crystals of iron were prepared by a strain anneal technique[18]. The starting material was MRC-VP (Materials Research Corporation) grade (99.95%) rod with an approximate diameter of 9.5 mm. The impurity content of a rod is presented in Table 1. These single crystal rods were cut in the Electric Discharge Machine (EDM) into disks, the thickness of which ranged from 0.3 to 1.2 mm. Tensile specimens were sliced along the rod axis and the final shape was produced with the EDM.

These specimens were mechanically polished with diamond paste and then 0.03 µm alumina slurry. Next they were chemically polished in a solution of 80% hydrogen peroxide (30% aqueous solution), 5% hydrofluoric acid (48% aqueous solution), and 15% water. The dimensions of the tensile specimens in the gage area were 11 × 3.0 mm and the range of thicknesses was between 0.4 and 1.1 mm. The polished samples were annealed and purified in the circulating ZrH_2 furnace at 1120 K for about 80 h. A thermodynamic argument was reported by Stein *et al.*[19] that the carbon, nitrogen, and oxygen contents should be almost undetectable after this treatment. More recently Meshii and coworkers[20] reported that the content of interstitial impurities such as C and N could be reduced substantially more than the O content.

Some of the disk-shaped specimens were bent over mandrels of several diameters at room temperature. The orientations of the disk specimens, the Schmid factors (i.e., the product of cosines used to determine the resolved shear stress along the slip direction)and the tensile axis will be presented in the Appendix. Deformation was carried also out by tension in a dry ice and methanol bath at 200 K with a strain rate 1 $\times 10^{-4}$/sec. Shortly after deforming, the specimens were quenched into liquid nitrogen.

In addition, strap shaped crystals of a different orientation were treated in the same manner were deformed both at 200 K and

INTRODUCTION

It has been recognized for some time that positrons can be trapped by dislocations before they annihilate, and that their lifetime[1-4] is similar to that of other defects such as a monovacancy.[5] Apparently the first observation of trapping in dislocations was made in 1964 by Dekhtyar *et al.*[1] who deformed a tin sample by hammering: this was followed in 1967 by Berko and Erskine[2] who studied the angular correlation of annealed and deformed aluminum and concluded that the positron would be localized on the dilatational side of an (edge) dislocation. This was before the introduction of the trapping model by Bergersen and Stott[6] and by Connors and West.[7] A number of studies followed. Doyama and Cotterill[3] listed in the 1979 Conference the lifetimes for annihilation in the bulk and in dislocations for a number of metals, and these are quite close to the currently accepted values. Most of the detailed studies which involve positrons interacting with a metal have been concentrated on vacancies and vacancy clusters. A number of studies have recently been published[8-12] in which the annihilation characteristics of cold-rolled metals are described in terms of the generic term, "dislocations"* and alternatively in the case of iron, in terms of carbon-vacancy interactions[13] and even in terms of an interaction with a self-interstitial by Frank *et al.*[14]. There has been little agreement on whether the positron is localized in the vicinity of the dislocation or in its core or by some other defect associated with a dislocation. This may reflect the general conviction best expressed by Siegel[15] that positrons will not readily bind to dislocations but only at jogs along them. It was reasoned that the lifetime in this trap should be similar to that of a vacancy. Doyama and Cotterill[3] were the first to argue that "once a positron arrives at the core of a dislocation, it diffuses very quickly [pipe diffusion] until it finds a vacancy attached to the dislocation or a jog of the dislocation, is trapped and annihilates there." There is little convincing evidence today that the diffusion of interstitial atoms such as carbon or hydrogen, is rapid along the dislocation core. In a recent paper, Tabata and Birnbaum[16,17] presented evidence of the effect of hydrogen on enhanced dislocation mobility. In the present research, the permeation of hydrogen through deformed iron specimens and the number of traps contained in the specimens will be presented in a subsequent paper. The number deduced will be found to be in good agreement with the number of dislocations determined here by positrons. Transmission Electron Microscope pictures taken of specimens deformed at 200 K show relatively straight dislocation lines aligned along <111> directions and emergent dislocations have been photographed with their associated etch pits. Thus the connection

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*By this phrase the authors mean a single trap component was fitted and labelled "due to a dislocation" when several types of dislocations were probably present.

DISLOCATION STUDIES ON DEFORMED SINGLE CRYSTALS OF HIGH PURITY IRON USING POSITRON ANNIHILATION: DETERMINATION OF DISLOCATION DENSITIES

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ABSTRACT

A new method for determining the total number of dislocations per unit area, as well as the fraction of screw and edge components, is discussed. Single crystal specimens which have been (a) bent over a mandrel, (b) cold rolled at room temperature, and (c) deformed in tension at 200 K, as well as iron whiskers, have been studied. Etch pits were studied in connection with this work and were found to be primarily of the <111> type. The density of emergent dislocations as determined by the etch pits was found to be smaller than the number determined by the positron annihilation technique.

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work are given.

1. Discussing Jogs versus Dislocations.

In 1979, Doyama and Cotterill[3] referring to their paper stated that "There is not much of an opening of ions at the core of a dislocations. Therefore positrons probably do not annihilate at the normal sites of the core of the dislocations." In their figure I, the arrangement of atoms on three consecutive {112} planes does not indicate much dilation around the screw dislocation. However, using anisotropic elasticity theory, an alternating three-fold dilatational field is indicated for a screw dislocation[41]. Although the dilatational field is 60 or 70% smaller than around an edge dislocation, it is still appreciable.

Doyama and Cotterill to make the point illustrate the constriction of dislocation ribbons associated with the formation of a jog, The drawing appears to be applicable to face centered cubic metals. However Argon and Moffat[53] have shown the atomic positions near an acute jog in a body-centered cubic metal. One can see appreciable "spaces" which occur that are similar to those along the edges of the ribbon.

Smedskjaer, Manninen, and Fluss[54] suggested that there is insufficient interaction between an (edge) dislocation and a "positron to trap the latter long enough to increase its life-time. Hence, it was more probable that the dislocation would serve as temporary storage and that diffusion would occur readily along or parallel to the core of the dislocation occurred rapidly until the positron came under the influence of a jog." They suggest that the jogs might be 100 to 1000 Burgers vector |b| lengths apart. The latter (jogs) would be expected to form by dislocation-dislocation interactions - mainly by one dislocation cutting across another: i.e, to form when the dislocation density becomes high. Cross slip could also produce this type of jog. While they presented a kinetic analysis to show that certain phenomena could be explained by means of their model, there has been very little direct evidence presented to support this concept.

Admittedly most of the positron studies have been carried out in the past after a substantial reduction in thick-ness has occurred, i,e., after 20 to 50% cold work. It would be very likely that complex dislocation arrangements would be observed with the electron microscope. In fact, there appears to be a paucity of confirmatory examination of specimens in the transmission electron microscope either before or after submitting these specimens to positron annihilation.

The positron may be "trapped" but not immobilized within the core of the dislocation. It does not escape, it merely finds a deeper trap somewhere along the dislocation at, for example, a jog. This implies a high positron diffusivity (or mobility) down the core. While it is well demonstrated that the diffusion of substitutional atoms is facilitated by pipe diffusion along a dislocation, there seems to be much less convincing

evidence for pipe diffusion of interstitial atoms (other than self-interstitials.) There is evidence that hydrogen is transported by moving dislocation during straining.[16,17,55].

Smedskjaer's calculations were based on a dislocation-positron binding energy of 0.1 eV. Snead *et al.* found the same value, 0.10 ± 0.05 eV, for the binding of a positron to a "dislocation" in Mo. However, the only value reported for iron is that by Van Brabander[31] which is 0.55 eV. Thus the conclusions about insufficiently strong trapping for Fe may be suspect. Nevertheless, because of the general conviction and "predictions" that (a) trapping would not occur in screw dislocations and (b) even if trapping were possible, the "t ap" would be so weak that it might only be detected at liquid helium temperatures. Thus it was felt that the possibility should be explored experimentally for iron at low temperature. Only low strain tensile deformations, either (a) controlled bending, or (b) cold rolling limited to a few percent were used to minimize the possibility of introducing a significant number of jogs.

Low and Turkalo[56] studied dislocation multiplication in silicon-iron single crystals using a transmission electron microscope. In specimens deformed by compression, at 1% the number of jogs in one cm length of screw dislocation was approximately 2 $\times 10^4$. This means that the average distance between jogs is about 0.5 µm, and the concentration of jogs in the specimen in which the density of dislocations is 5 $\times$ $10^{13}/m^2$ reduces to 1.2 $\times$ 10^{-9} per Fe atom. This frequency of jog occurrence is very similar to the number which Smedskjaer *et al,* [54] used, namely a jog every hundred |B| along the dislocation. Even if the jog is assumed to have 10 times higher trapping capability than a monovacancy this concentration is too small to give a significant signal i.e., to be detected by the positron annihilation equipment. The lower limit for detection of vacancy concentration is of the order of 0.1 ppm [57] with currently available equipment.

The transmission electron microscope pictures taken by Kimura and Kimura.[59] of lightly deformed single-crystal tensile deformations of similarly oriented iron specimens at 200 K show only relatively simple arrangements of dislocations. Electron micrographs to presented by the present authors in a later paper are very similar. One of their typical stress-strain curves is presented in Figure 6 along with two of ours. The principal defects they observed during small deformations are edge dislocation initially and then primarily non-kinky screw dislocations of the <111> type. According to Keh and Weissmann[50] the distribution of dislocations is relatively uniform when formed at -75 C and -135 C: cells are not formed below 16% strain. They arrived at the conclusion that primarily straight screw dislocations of the <111> type were formed up to 13% strain.

Ikeda[51] studied the dislocation distribution in iron single crystals at 200 K and room temperature. While the transmission electron micrographs were different depending on the orientation of the plane where the micrographs were taken,

micrographs taken in the specimens deformed at 200 K by [the present or other] authors are similar to the Figure 3 (a) and (b) of Ikeda which shows short and straight screw dislocations and edge type loops at low strains(2-6%).

Solomon and McMahon[37] determined the fraction of edge and screw dislocations which were formed in iron single crystals at 77 K. The fraction varied from 40 to 70 percent screw component according to the stress level.

2. Discussing Two Lifetimes for a Dislocation.

In a number of earlier studies on deformed iron basically two lifetimes were found; one for annihilation in the bulk and the other for a defect which was most likely a dislocation. Doyama and Cotterill[3] reported 117 and 169 psec, Cao Chuen *et al.*[30] gave 111 and 162 psec, Van Brabander *et al.*[31] reported 108 and 167 psec,and Nielsen *et al.*[58] reported 165 psec for a steel.

When we first the authors had observed the lifetime of 165 psec a significant number of times with carefully bent specimens, (and before we had found the consistent set of lifetimes just cited) Vehanen[24] suggested that this value might well be due to a mixture of traps one of which was with 157 psec, which the Finnish group had assigned to (undifferentiated) dislocations, with 175 psec assigned as the lifetime of a monovacancy. Subsequent work has failed to verify this suggestion.

In addition, Kuramoto *et al.*[49] carried out very similar deformation of iron single crystals and made positron annihilation measurements. They were convinced a *priori* that dislocations would be very unlikely to be detectable. They annealed their low-temperature deformed specimens for several hours at room temperature. They reported their results only in terms of a fixed trap with a lifetime of 175 psec and vacancy clusters. This was probably because Cotterill and Doyama [3] had expressed the opinion that that there was little likelihood trapping in the dislocation and the lifetime of a positron in a jog might be similar to that of a monovacancy. They [43] did observe a second lifetime in the vicinity of 350 psec.

Our data were reanalyzed using 175 psec as a fixed lifetime, but we were unable to obtain statistical justification for forcing this value rather than using 165 psec. We did a number of further analyses since we had observed a single-trap lifetime of 155 psec with cold-rolled specimens. The contrary hypothesis occurred to us, namely that since cold rolling takes place with a more complex stress state at various parts of the (single crystal) sheet, that 155 psec might be due to a mixture of traps. Indeed 165 psec was found together with 143 psec. No strong evidence was obtained for 175 psec after examining a number of carefully deformed specimens . In each case, the X^2/ν measure of statistical fit was of significantly lower quality.

The trapping by a mixture of edge and screw dislocations can, in principle, be analyzed with a two trap model since the two lifetimes are sufficiently well separated. The number of dislocations with either type of component can thus be obtained. The lifetimes of 114 and 165 psec were fixed and the data were reanalyzed. The values of the lifetime determined in the two ways are presented below together with their X^2/ν values in Table V.

In these runs, it was difficult to fit the second trap. Nevertheless, the corresponding values of X^2/ν in most cases were not significantly different as they would have been if an additional trap was not warranted.

There is another observation of 142 psec for iron; it has been reported by Vehanen[61] for a C-vacancy pair. However, this value is not the lifetime of a single type of a trap but rather a "mean" lifetime which comes from assuming only one trapping state, It was resolved by these authors into one component with a lifetime of 160 psec.

3. Discussing The Specific Trapping Rates

Cotterill *et al.*[59] suggested that the cross section for trapping a positron in a defect could be obtained using the equation

$$\mu = \sigma \langle v \rangle \rho \qquad (5)$$

where μ is the specific trapping rate, σ is the cross section, $\langle v \rangle$ is the mean velocity of a thermalized positron and ρ is the defect density. If one knows the cross section associated with a specific type of trap, μ_T can be calculated by equation (5).

Recently a Chinese group calculated the specific trapping rate by this equation and calculated the density of defects in an iron specimen with a cold reduction in area of 60%. It is reasonable to assume that a mixture of dislocations and vacancies resulted from this heavy reduction. In determining the number of dislocations per unit area, they used the σ value of 1×10^{-15}. This unfortunately is the σ value Snead *et al.*[60] reported for a vacancy in iron. Vehanen *et al.*[61] obtained a specific trapping rate from positron work on irradiated iron crystals of 1.1×10^{15}/sec. Thus the specific trapping rate Cao *et al.*[30] presented namely 8.99×10^{14}/sec was really for a vacancy.

Dlubek *et al.* [62] compared the specific trapping rates for dislocations and vacancies in Ni. They converted the units of the specific trapping rate from [cm²/sec] to [sec⁻¹] by multiplying by the Burgers vector and dividing by the volume of the radius of the atom. The specific trapping rates of dislocations and vacancies they obtained were 2.9×10^{15}/sec and 2.2×10^{15}/sec respectively. The specific trapping rate was slightly higher in a dislocation than in a vacancy and they remarked that

the same was true for copper. If the specific trapping rates obtained in the present study were converted to the same basis as Dlubek *et al.*[62] used, they would be 1.07×10^{15} for a screw and 1.47×10^{15} for an edge dislocation. For this calculation we used the Burgers vector $|b| = \frac{1}{2} \langle 111 \rangle$. Note that these specific trapping rates are larger than the two values reported for a vacancy namely, 0.899 and 1.1×10^{15}/sec. There is additional evidence that our trapping rates are reasonable. Dlubek *et al.* [62] obtained 1.2×10^{-4} m^2/sec for the specific trapping rate of a dislocation in Ni. McKee *et al.*[63] obtained 2.9×10^{15}/sec for this type of defect in Cu.

4. Discussing the Temperature Independence of the Trapping Rate

In determining the dislocation densities from the measured values of κ_T at 300 and 200 K, the authors assumed that the two specific trapping rate μ_E and μ_B were not temperature dependent. A shallow trap would probably behave differently and thermal detrapping might occur at these low temperatures. Although the trap for an edge might be deeper than for a screw dislocation, there is no direct experimental data to bring to light on this question raised because of two different types of dislocation traps. Thus we have relied on several recent papers which show that the trapping mechanism for positrons is essentially independent of temperature at room temperature and below. Bergersen and McMullen[64] gave a theoretical explanation. Dlubek *et al.*[62] experimentally verified this for nickel and Rice-Evans *et al.*[66,67] confirmed it for copper.

There is one indirect way to attempt to address this question. The R parameter should vary with temperature if there would be a change in the population of traps during an experiment. It is interesting that the R parameter was found by Cao *et al.*[30] to be essentially constant for cold-rolled iron up to about 500 C. In discussing the R parameter, Mantl and Triftshauser[52] noted that the trapping constant for vacancies in aluminum and copper was temperature independent below room temperature. They show that for aluminum R is constant over the temperature range 4 to 500 K. The probability of trapping in a given defect state T is

$$f_T = \mu_T C_T \;/\; (\lambda_T + \mu_T C_T) \tag{6}$$

then one can show from the linear combination of peak values that

$$C_T \begin{bmatrix} \mu_T \\ \text{--} \\ \lambda_{*} \end{bmatrix} = \frac{(P \;\; - P_B)}{(P_T - P \;\;)} \tag{7}$$

Since one knows the annihilation rate λ_T, it is possible to determine the specific trapping rate μ_T from the intercept on a log-log plot of the right hand side versus the trap concentration C_T as long as that one type of trap dominates the annihilation process. They show that such a log-log plot does give a

tion process. They show that such a log-log plot does give a straight line with unit slope for electron-irradiated copper. They obtained a specific trapping rate of $0.425 \pm 0.08 \times 10^{15}$ sec^{-1}. This is indeed much smaller than the value given by McKee *et al.* above for a dislocation in copper.

5. Related Work

Byrne and his coworkers[35] deduced the dislocation content of a heat-treated eutectoid steel from x-ray line shapes, and measured the Doppler broadening P/W parameter on the same samples. They concluded that the positrons were annihilating in dislocations within the ferrite grains, It is interesting that they found P/W to yield a straight line when plotted against the dislocation densities in the range of $1 \times 10^{14}/m^2$. Their value of the specific trapping rate μ_T was 1.5×10^{15}/sec.

Xiong-Liang-Yue[47] studied the isothermal recrystallization of pure iron which had been rolled to 40% of the original thickness, The change in the dislocation concentration at 500 C was linearly correlated with the volume fraction of newly formed and hence "dislocation -free" grains. There was no change in the R parameter during recrystallization suggesting that there was only one type of positron trap - an undetermined and probably mixed dislocation - involved.

CONCLUSIONS

The consistent behaviour of positron annihilation trapping in a fairly large range of deformation experiments on single crystals of high purity iron has been presented.It has been shown that it is possible to determine the number of screw and edge dislocations per unit area by a combination of the two types of positron annihilation measurements, namely positron lifetime spectra and line-shape analysis of the Doppler broadened γ ray radiation. The number of traps is in good agreement with the density of dislocations revealed by etch pits. Separate specific trapping rates have been determined and compared with similar results which have been published elsewhere.

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Appendix A

The orientations of various potential slip planes with respect to the tensile direction are shown in Figure 14. The appropriate Schmid factors are listed in the accompanying table

Table VI. The Schmid Factors of Slip Systems

in Iron Tensile Specimens

Slip Direction	Slip Plane	Schmid Factor Crystal #1	Schmid Factor Crystal #2
[111]	(101)	0.482	0.495
	(211)	0.461	0.433
	(112)	0.383	0.422
[111]	(101)	0.464	0.485
	(112)	0.380	0.429
	(211)	0.417	0.406

It can be seen that the slip systems in crystal 1 are more nearly parallel to the wide surface of the tensile specimen. This has an appreciable influence on the dislocations which can remain in the specimen after deformation.

APPENDIX B

A few additional trials at fitting the lifetime data with different choices of the input parameters.

TABLE VII, Effect of the Number of Traps Assumed on the Chi Square Results For Iron Single Crystals Deformed at 200 K

One Trap τ_T	One Trap X^2	Two Floating Traps $\tau_T(1)$	Two Floating Traps $\tau_T(2)$	Two Floating Traps X^2	Two Traps, One Fixed $\tau_T(1)$	Two Traps, One Fixed Fixed	Two Traps, One Fixed X^2
154±3	1.218	147±5	159±7	1.210	145±3	165	1.234
147±4	1.172	139±4	166±13	1.203	138±3	165	1.196
155±5	1.180	145±6	161±9	1.177	144±4	165	1.172
Average 152±5		144±10	162±9		142±4	165	

The values of X^2/ν are less sensitive to the choices than had been anticipated. Thus we have adopted the position that the inclusion of the short lifetime or 142 psec is justified since the inclusion of another trap did not worsen the fitting and a more consistent interpretation.

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TABLE I Chemical Analysis of the Starting Material

MRC-VP grade Iron as reported by

the Materials Research Corporation.

(in wt. ppm)

H	C	N	O	S	Mg	Si	Al	Ca
<1	18	<1	33	40	<10	50	60	<10

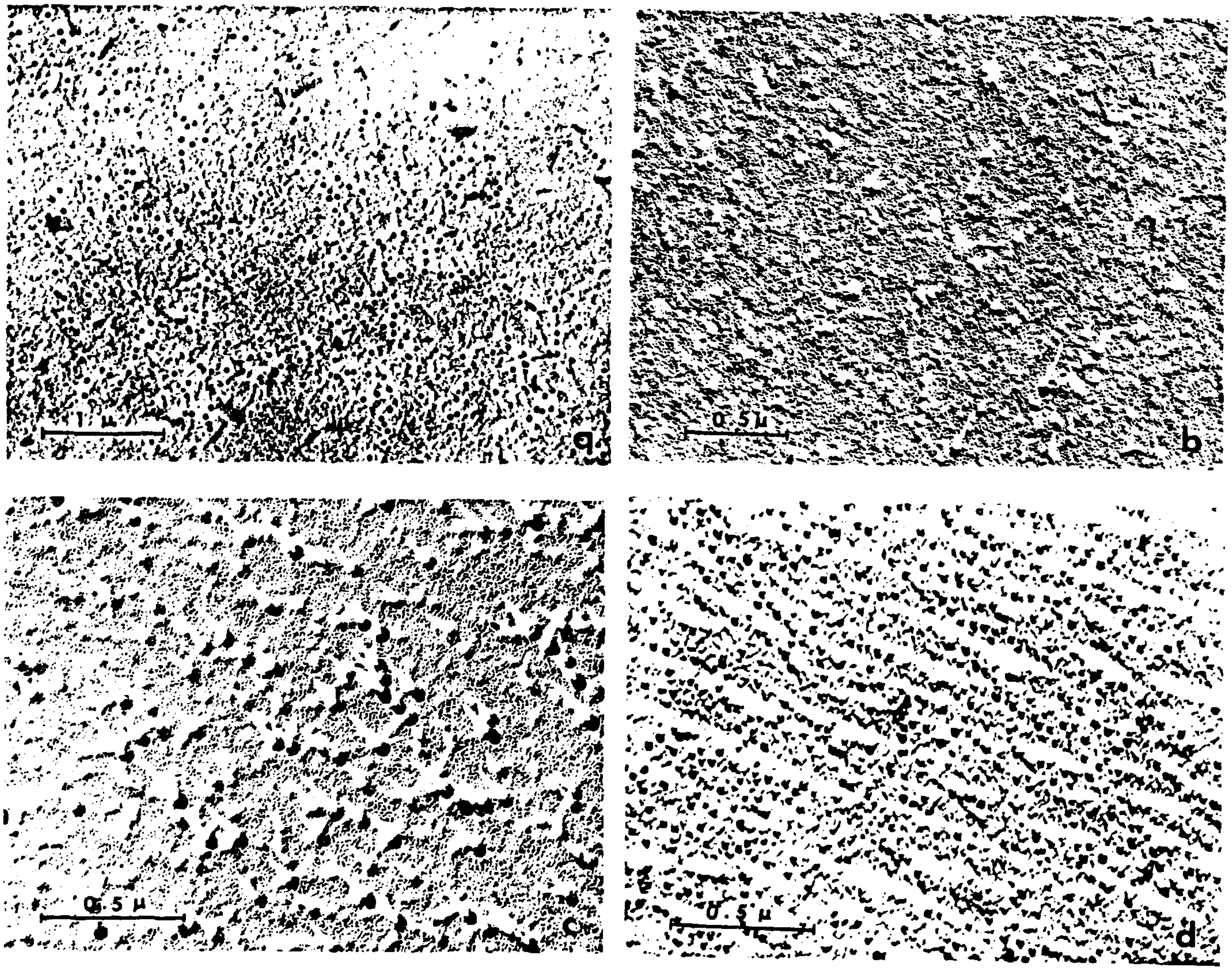
Ni	Cu	Ti	Cr	Mn	Ag	Sn	Pb	Others
<10	30	<10	30	30	<5	<30	<30	N D

TABLE II. Dislocation Densities in Iron Crystal 1

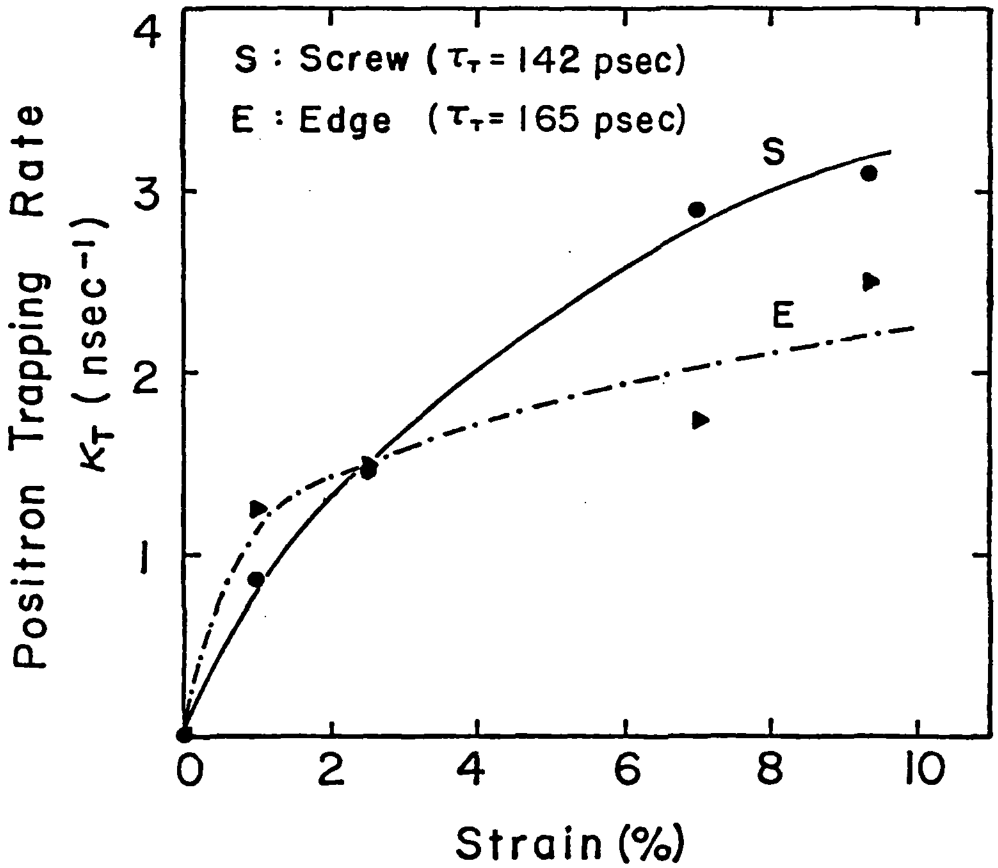
Deformed at 200 K which were obtained

from Etch Pit Counts

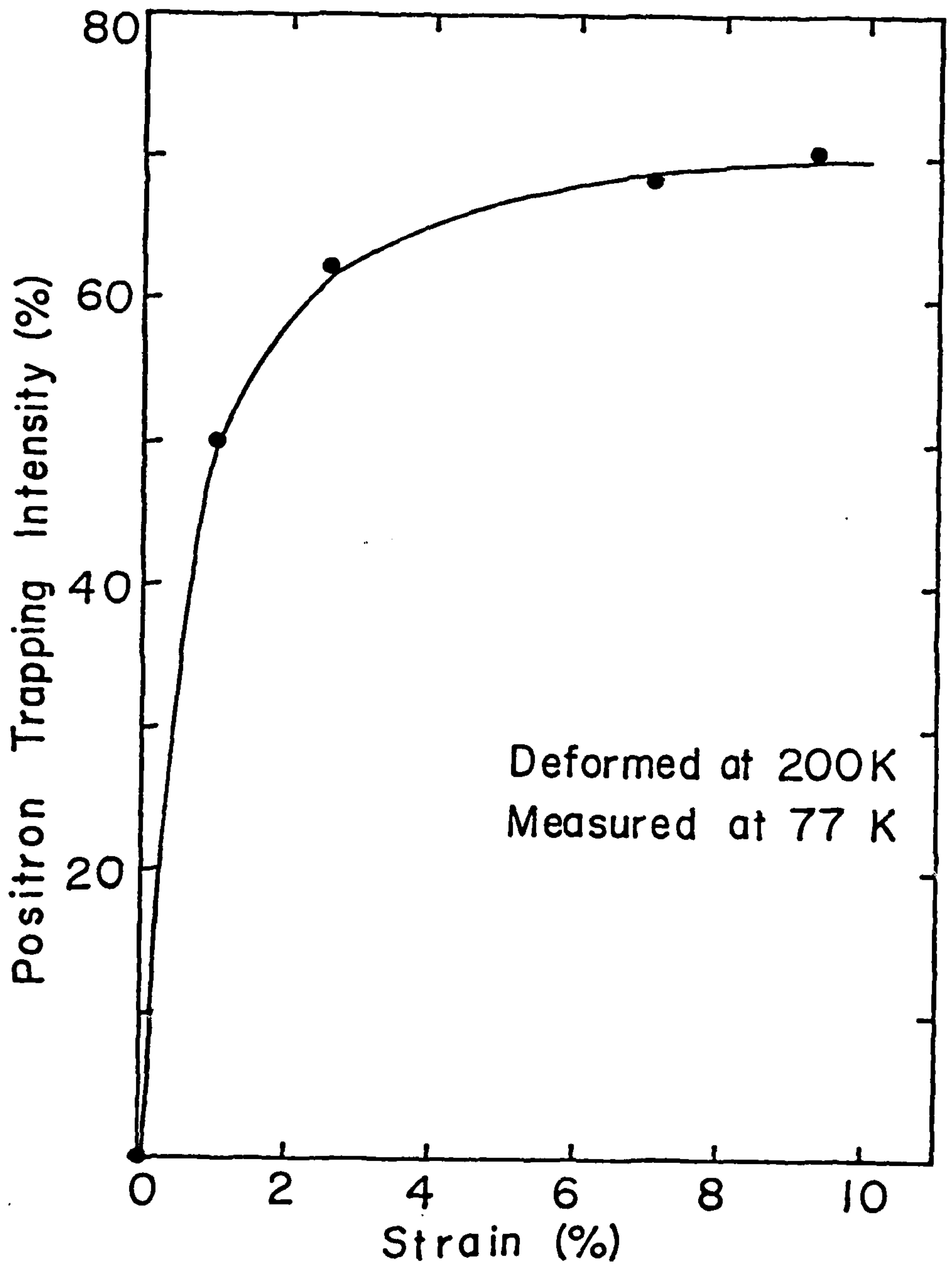
True Strain (Percent)	Dislocation Density ($\times 10^{13}/m^2$)
1	2
2.5	5
7.0	7
9.3	9



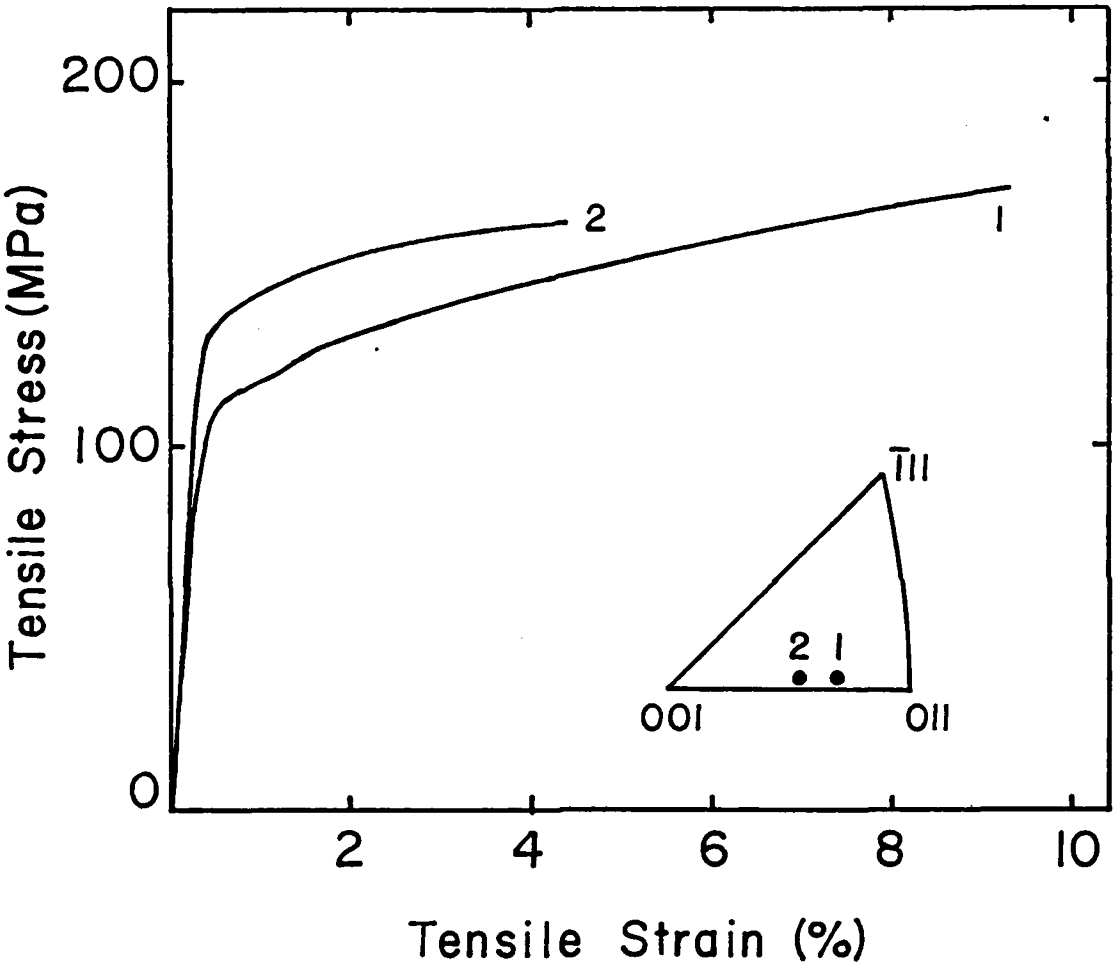
1 μ
a
0.5 μ
b
0.5 μ
c
0.5 μ
d



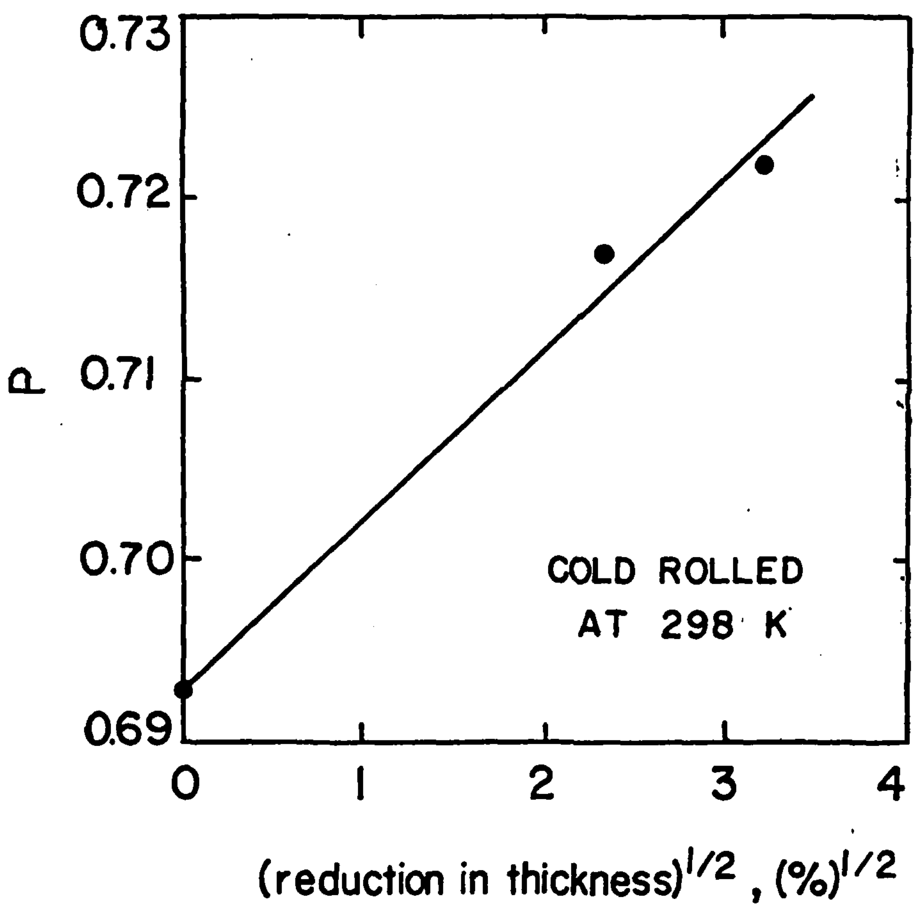
S : Screw (τ_T = 142 psec)
E : Edge (τ_T = 165 psec)
S
E
Positron Trapping Rate
κ_T ($nsec^{-1}$)
Strain (%)
0
2
4
6
8
10
1
2
3
4



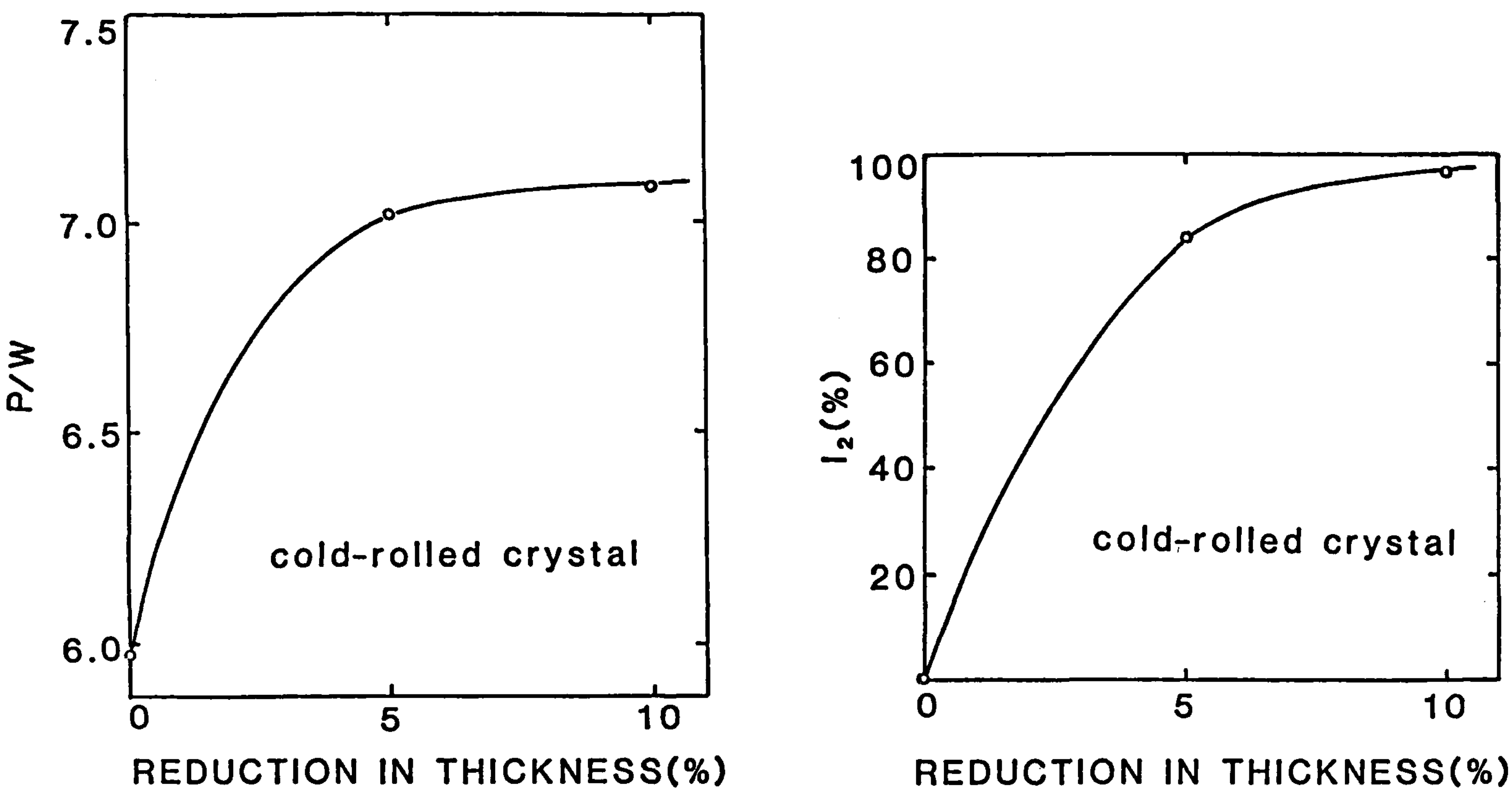
80
60
40
20
0
2
4
6
8
10
Positron Trapping Intensity (%)
Strain (%)
Deformed at 200K
Measured at 77 K



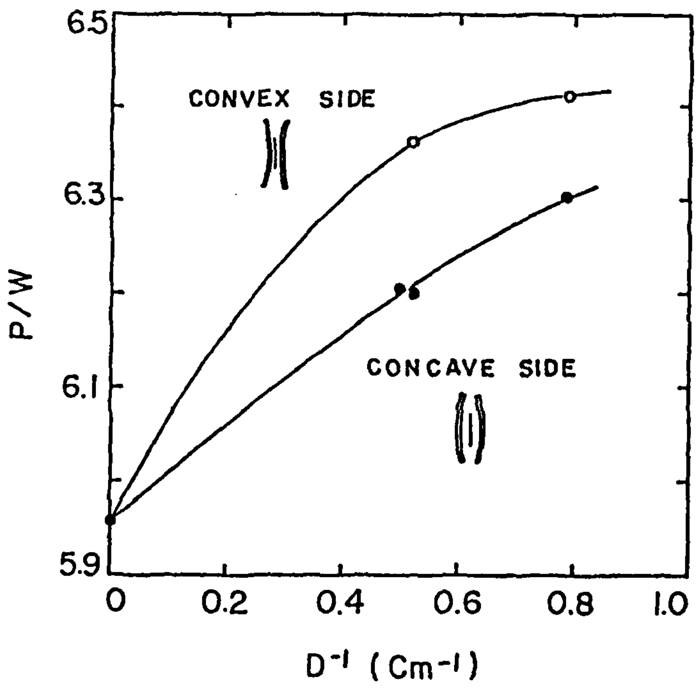
200
100
0
Tensile Stress (MPa)
2
1
$\bar{1}11$
2 1
001
011
2
4
6
8
10
Tensile Strain (%)



0.73
0.72
P
0.71
0.70
0.69
0
1
2
3
4
COLD ROLLED
AT 298 K
(reduction in thickness)$^{1/2}$, (%)$^{1/2}$

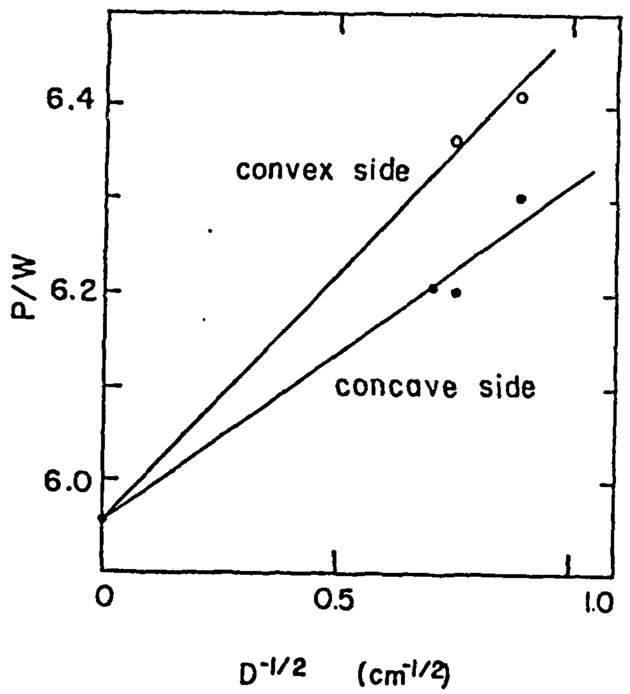

7.5
7.0
6.5
6.0
P/W
cold-rolled crystal
0
5
10
REDUCTION IN THICKNESS(%)
100
80
60
40
20
I_2(%)
cold-rolled crystal
0
5
10
REDUCTION IN THICKNESS(%)

(a) (b)



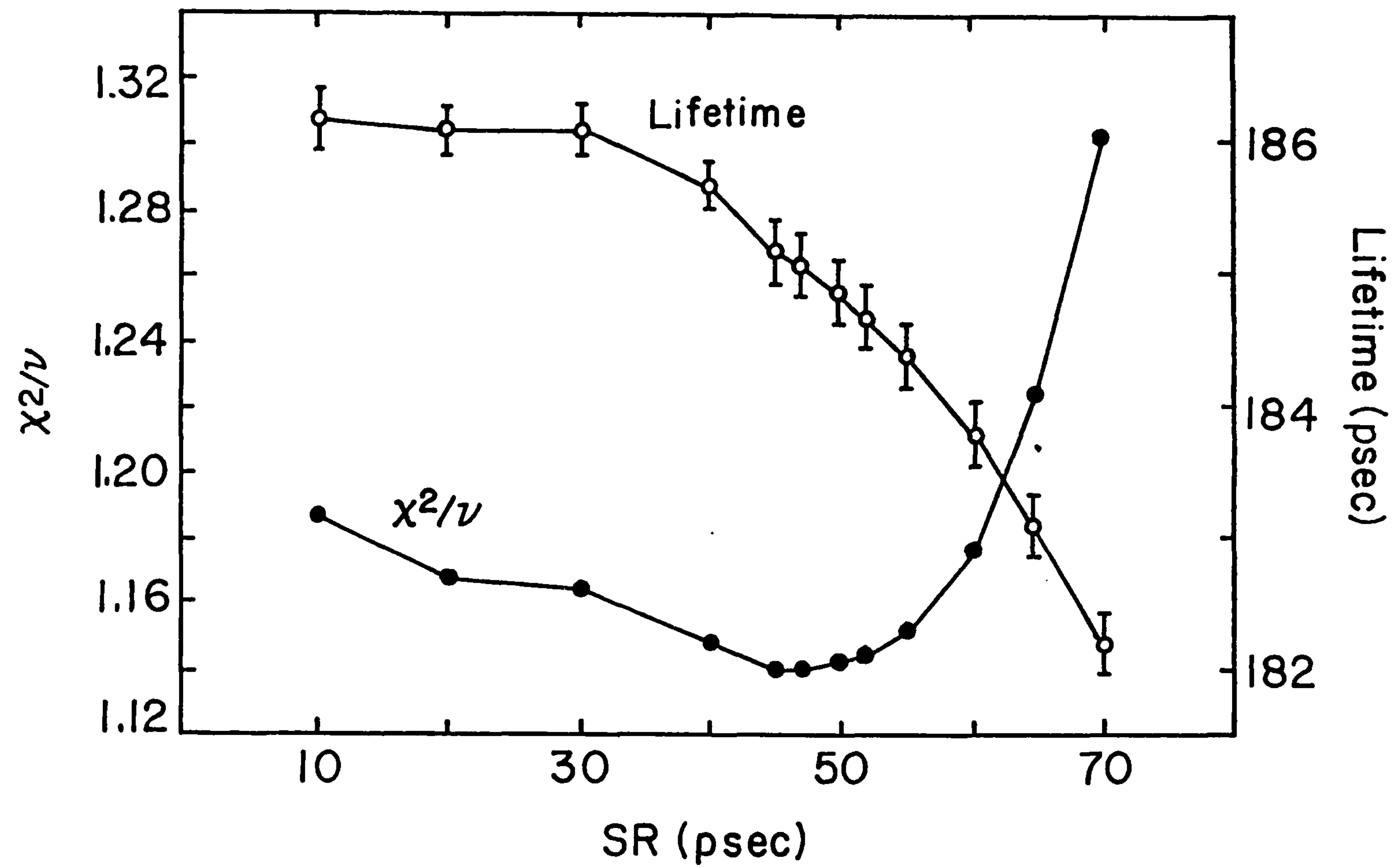
6.5
6.3
6.1
5.9
P/W
CONVEX SIDE
CONCAVE SIDE
0
0.2
0.4
0.6
0.8
1.0
D^-1 (Cm-1)

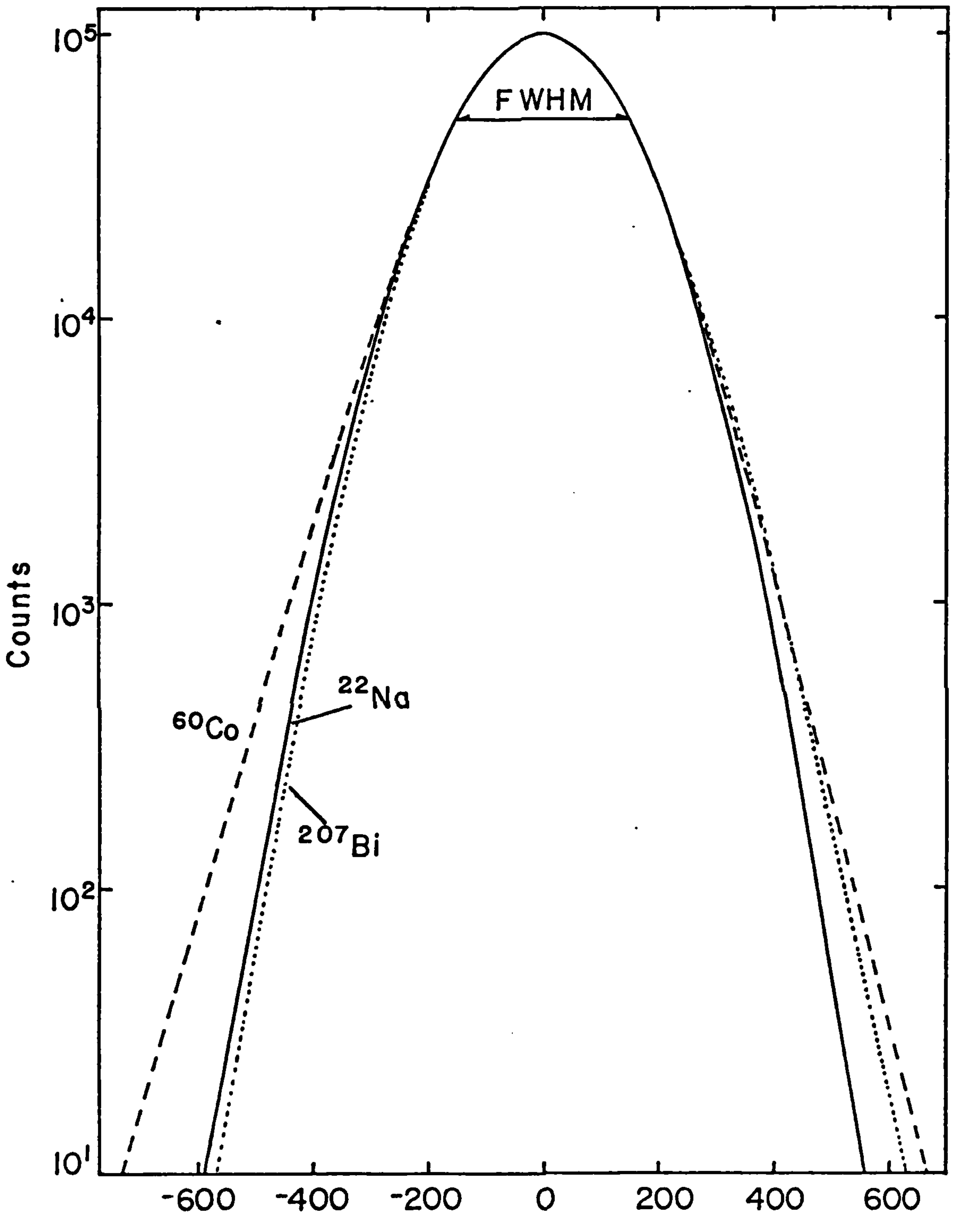
(a)



6.4
6.2
6.0
P/W
convex side
concave side
0
0.5
1.0
D^-1/2 (cm^-1/2)

(b)


Lifetime
χ²/ν
1.32
1.28
1.24
1.20
1.16
1.12
186
184
182
Lifetime (psec)
10
30
50
70
SR (psec)



10^5
10^4
10^3
10^2
10^1
Counts
FWHM
^{60}Co
^{22}Na
^{207}Bi
-600
-400
-200
0
200
400
600
Time (psec)

Fig. 12. The R parameter as a function of a tensile strain in single crystals of iron deformed at 200 K, where the solid square is for Crystal #2 which was annealed at room temperature for 48 hours. All measurements were done at 77 K.

Fig. 13. The R parameter plotted as a function of the inverse of the bending mandrell diameter of iron single crystals. Open marks represent the tension(convex) side and filled marks the compression(concave) side of the bent specimen.

Fig. 14. The orientation of iron single crystals and schematic drawing of the potential slip systems in the tensile specimens with relation to the direction of applied stress. Orientations 3-6 represents the surface normal of bent and cold rolled specimens.

FIGURE CAPTIONS

Fig. 1. Measured resolution function obtained with ^{60}Co and the fitted prompt curves measured with ^{22}Na and ^{207}Bi.

Fig. 2. The variation of the lifetime and X^2/ν as a function of the right hand slope of the resolution function(SR) for ^{207}Bi.

Fig. 3. Doppler-broadening lineshape parameter for bent iron crystals: (a) P/W parameter as a function of the curvature (reciprocal of the mandrel diameter), (b) the same parameters as a function of the square root of the curvature. The error bar is the same size as the marks. Solid lines are guides to the eye.

Fig. 4. Positron annihilation measurements on cold-rolled iron: (a) The peak-to-wing (P/W) parameter of Doppler broadening, (b) The fraction of the positron annihilating in the trap with the lifetime 155 psec. The error bar is the same size as the marks. Solid lines are guides to the eye.

Fig. 5. The positron annihilation Doppler broadening peak P parameter for cold rolled iron as a function of the square root of the reduction in thickness. The error bars are the same size as the points plotted. Solid lines are guides to the eye.

Fig. 6. Stress-strain curves of iron single crystals deformed in tension at 200 K.

Fig. 7. The total fraction of the positrons annihilating in traps at 77 K of single crystals #1 deformed in tension at 200 K.

Fig. 8. Positron trapping rates at 200 K in edge and screw dislocations in single crystal #1 deformed at 200 K in tension.

Fig. 9. Transmission electron micrographs of replicas of the etch pits developed on the surface of iron single crystal #1 deformed in tension at 200 K for increasing amounts. (a) ϵ= 1.0% strain, (b) ϵ= 2.5%, (c) ϵ= 7% and (d) ϵ= 9.3%.

Fig. 10. Dislocation density in iron single crystal #1, which was deformed in tension at 200 K. Solid circles represents the total dislocation density as measured by positron annihilation and open circles are the values from the etch pit measurements. Triangles and squares represent the edge and the screw components measured with positron annihilation.

Fig. 11. Dislocation density as a function of the true strain measured by various methods. (from Yamakawa *et al.*[48])

TABLE V. Results of Determining the Positron Lifetime with One and Two Trap Models on Specimens Deformed at 200 K

One Trap*			Two Traps**		
τ_T (psec)	χ^2/ν	I_2 (%)	τ_T (psec)	χ^2/ν	I_3 (%)
154 ± 3	1.218	78	145 ± 3	1.234	58
153 ± 5	1.180	76	144 ± 4	1.172	56
150 ± 13	1.189	68	140 ± 33	1.189	40
144 ± 14	1.169	72	141 ± 7	1.177	33
148 ± 8	1.514	76	144 ± 10	1.536	49
141 ± 7	1.191	86	137 ± 11	1.175	71
148 ± 8	1.402	72	140 ± 9	1.417	49
157 ± 2	1.553	71	150 ± 8	1.529	43
154 ± 6	1.425	68	147 ± 10	1.426	44
148 ± 10	1.175	77	135 ± 13	1.178	69

* the lifetime for annihilation in the bulk was fixed

** both 114 and 165 psec lifetimes were fixed

Table IV Calculated Total Density of Dislocations in Bent Single Crystals of Iron (in Units of $10^{13}/m^{-2}$)

Diameter of Mandrel	Concave Side	Convex Side	Diff..
50.8 mm	1.0	1.5	0.5
20.2	1.3	2.5	0.8
19.1	2.0	3.3	1.3
12.7	2.9	3.9	1.0

(Lifetime observed was 165 ± 5 psec)

TABLE III. Specific Trapping Rates and Dislocation Density
in Cold Rolled Iron Single Crystals

Deformation %	Total Density[1] m^{-2}	μ_d [2] m^2/sec	Total Density[3] Calculated m^{-2}	Estimated Screw Density m^{-2}	Estimated Edge Density m^{-2}	μ_s [4] m^2/sec	μ_e [5] m^2/sec
5	1.5×10^{14}	6.8×10^{-5}	2.1×10^{14}	0.9×10^{14}	0.6×10^{14}	6.1×10^{-5}	7.8×10^{-5}
10	5.0×10^{14}	4.7×10^{-5}	5.6×10^{14}	3.3×10^{14}	1.7×10^{14}	4.0×10^{-5}	6.2×10^{-5}
						5.1×10^{-5}	7.0×10^{-5}

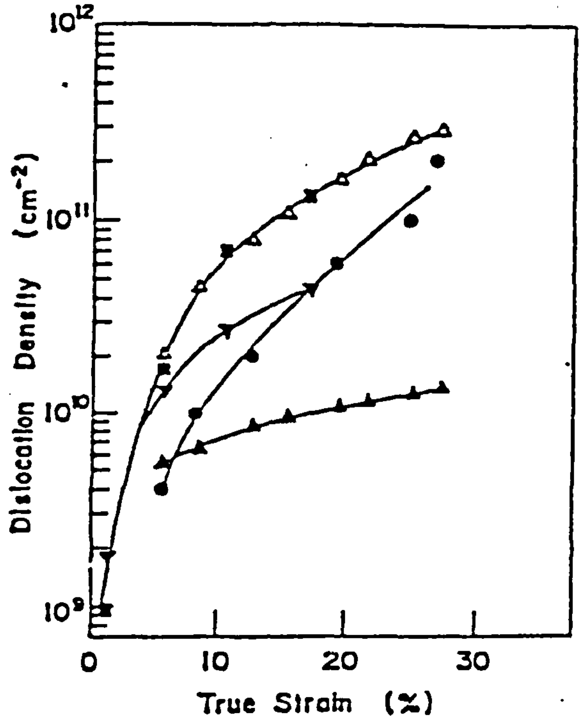
1. Reported values of Yamakawa *et al.* (Ref. 48).

2. "Mean" specific trapping rate for dislocation.

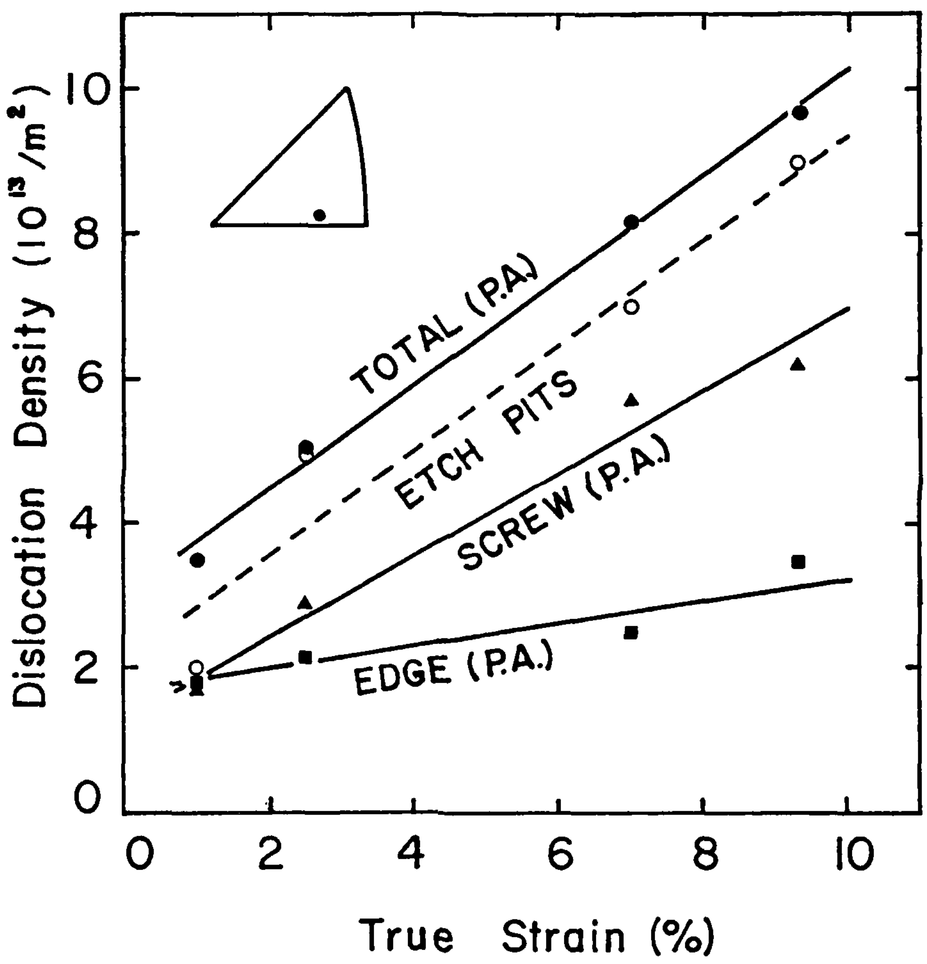
3. Total dislocation density calculated using Doppler broadening data and μ_d.

4. The specific trapping rate in the screw component.

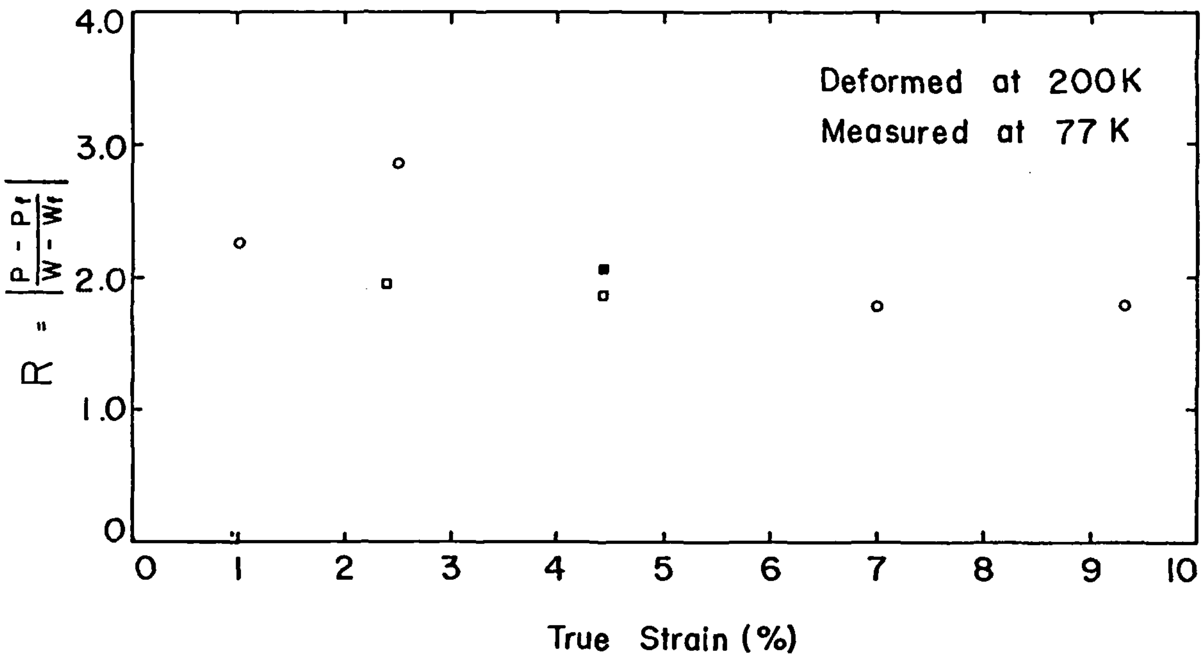
5. The specific trapping rate in the edge component.



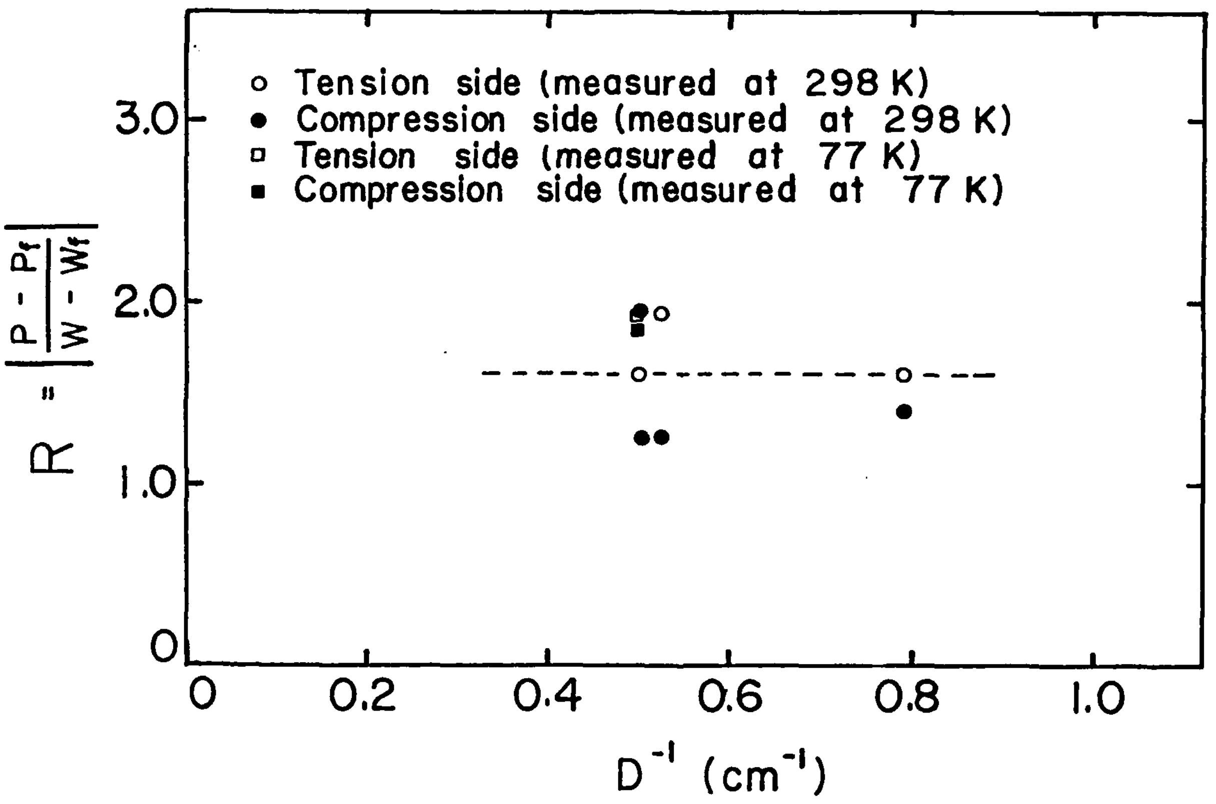
● hydrogen content
▲ X-ray diffraction profile microbeam
▼ X-ray diffraction profile
△ calibration of (▲) values
■ observation with electron microscope



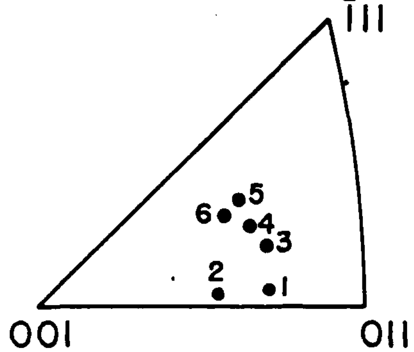
Dislocation Density ($10^{13}/m^2$)
True Strain (%)
TOTAL (P.A.)
ETCH PITS
SCREW (P.A.)
EDGE (P.A.)
0
2
4
6
8
10

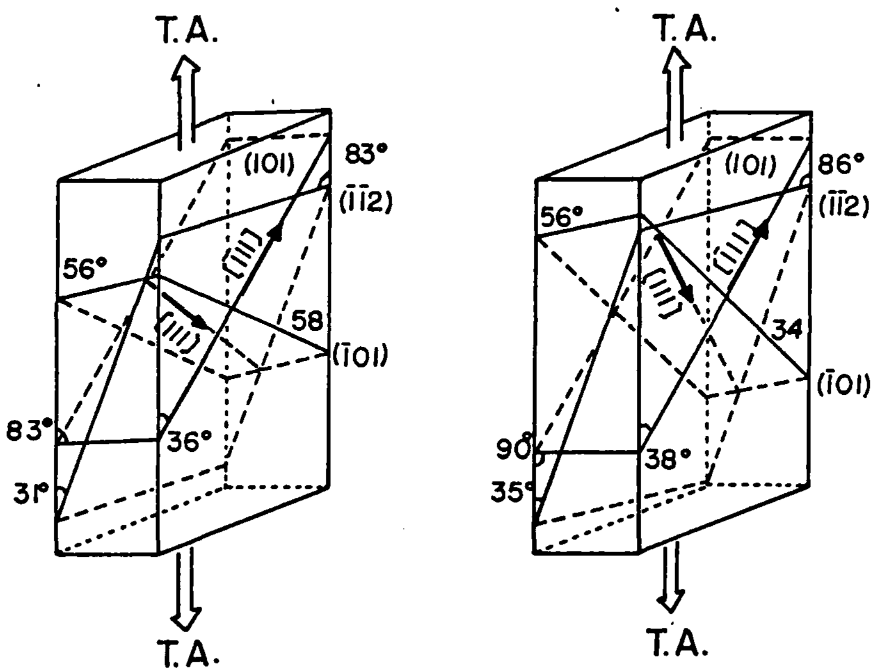


4.0
3.0
2.0
1.0
0
0
1
2
3
4
5
6
7
8
9
10
Deformed at 200K
Measured at 77 K
$R = \left|\frac{P - P_f}{W - W_f}\right|$
True Strain (%)



Tension side (measured at 298 K)
Compression side (measured at 298 K)
Tension side (measured at 77 K)
Compression side (measured at 77 K)
$R = \left|\frac{P - P_f}{W - W_f}\right|$
3.0
2.0
1.0
0
0
0.2
0.4
0.6
0.8
1.0
D^{-1} (cm^{-1})





Crystal 1 Crystal 2

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19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

N-atom clusters 3
Hartree-Fock calculations 2
Metal-substrate interactions 1
Cluster-cluster interactions 1

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Need for verifying or "calibrating" calculational methods by doing the same 100 atom cluster, by essentially all of the approximate, semi-empirical and ad-initio methods. Test cases are suggested.

To be published as a D.O.E. Panel Report.

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Report of the Panel on

See Section 2.2 of

THEORY AND COMPUTER SIMULATION OF MATERIALS
STRUCTURES AND IMPERFECTIONS

August 6-10, 1984
Michigan Technological University
Houghton, MI 49931

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complexity of materials, e.g., quaternary oxides and feldspar structured minerals.

(ii) Calculation of free energies in addition to energies, thereby enabling predictions of phase diagrams to be made. This latter area would be of particular value to ceramists and geophysicists.

(iii) Calculations of energies and entropies of solid state reactions.

Probably the most critical factor in allowing these developments to occur will concern interatomic potentials. In many systems, a realistic inclusion of many body terms in the potential is essential.

In addition, improved parameterization of potential models is needed. These improvements will require (a) an increased quantity and quality of crystal data to assist the construction of empirical and semi-empirical potentials, and (b) input from quantum mechanical calculation.

The second main need is for increased computer power if large numbers of highly complex materials are to be handled. Good simulations of, for example, a zeolite structure, may take about five CPU hours on the CRAY-1/S. Computer resources on this scale are required if the scope of the field is to be exploited. The potential applications to realistic calculations on large complex solids are, however, considerable.

2.2 The 100-Atom Cluster J. T. Waber

The problem of the equilibrium crystal structure adopted by a group of N atoms, where N is of order 100, poses a feasible test

problem for the comparison of various classical and quantum mechanical methods and also for the comparative testing of various force laws and ad-hoc potentials. In addition, such studies provide tests of the efficiency of the various strategies and their implementation.

Small clusters, known to possess interesting properties, are used in applications ranging from catlysis (Bond, 1962) to underwater fuels and explosives (Miller, 1984). Furthermore, recent experimental evidence indicates the possibility of magic number clusters (Sattler, 1982; Muhlbach et al., 1982), certain numbers of atoms produce clusters with binding energy per atom significantly greater than others. The initial indication is that these magic numbers are associated with configurations of several tetrahedra stacked together, forming a closed outer shell. Systems studied include Pb_n, Ar_n, $(CO_2)_n$, $(SF_6)_n$.

The theorist might well address the following questions: Can we find evidence for enhanced stabilities for certain magic numbers, how does the stability depend upon symmetry of the cluster, the atomic constituents of the cluster, the valency of the constituents, or the temperature of the cluster? It is anticipated that metallic clusters might differ significantly from covalent structures such as γ-Al_2O_3.

Doubtless this is a problem in which various theoretical methods can confront one another, as well as experiment, and is singled out for this reason. Clearly molecular dynamics methods, Monte Carlo methods using Lennard-Jones, Morse or Buckingham potentials can trivially deal with free-space bounded 100-atom

clusters of rare gas or metal atoms. Furthermore, using what amounts to lattice statics approaches, such systems could be studied by a variety of quantum mechanical techniques. It should be possible to study at least a few systems of this size using semi-empirical approaches (such as CNDO, MINDO, etc,) or ab initio methodology (local-density methods, Hartree-Fock methods, etc.) This latter suggestion is not fatuous, as some Hartree-Fock level studies have been made on systems containing at least 65 atoms (e.g., Beck and Kunz, 1984) on a VAX 11/750 computer. The development and calibration of many techniques could well be established using studies of N atom, rare gas or metal atom clusters. Clearly studies, if successful on simple systems, can be directly extended to more complex systems such as ionic compounds and covalent systems.

Having dealt successfully with the finite cluster in free space, one's attention is rapidly drawn to related problems of technological importance. These would be (a) the communication between such a cluster, or even a smaller one, and a support or substrate, and (b) the interaction between such clusters on a substrate. They are of considerable importance, for example, in areas such as heterogeneous catalysis or cladding. In the case of a catalyst, the optimum active site is often a few, to a few tens, of metal or mixed metal atoms bonded to a silica, alumina or silica-alumina substrate (Sinfelt, 1973).

Time-lapsed pictures have been taken by Pashley (1963, 1965, 1966) of the in-situ deposition of gold onto molybdenite (MoS_2) by use of electron microscopy. Normal faceted crystals were seen to

have separations comparable in size to a crystal dimension and of order 1 μm for low coverage. As deposition increased, however, necks developed between some of the islands, and the necks developed rapidly in width. Finally, the crystallites coalesced into a new faceted crystal in the region of the neck. Possibly elastic strain fields in the substrate are the means of communication between crystallites. Systems such as this should be amenable to modeling now (or as computer capabilities develop) by most of the techniques described for the 100 atom cluster. This is provided the total system size is limited, or the more time-consuming techniques were applied only to fragments of the problem. Clearly, some of the fine particles bonded to substrates used for catalysis are also amenable, albeit barely, to study by any of the techniques described.

2.3 Interfaces

Models of interfaces have provided valuable insight into many of the physical processes that occur at the boundaries of bulk phases, including crystal growth, catalysis, sputtering, sintering and abrasion. Most atomic-scale models have been derived from either the Ising model or systems of particles interacting by pairwise additive or triplet potentials. Simulations are usually performed using Monte Carlo or, in the latter case, molecular dynamics techniques. The Ising model is most appropriate for studies of systems that include extended structures, such as adatom clusters, or arrays of steps on close-packed crystal faces. The Ising model exhibits a number of crystal growth mechanisms,

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AD-A158 437 EFFECT OF INTERSTITIALS ON THE TRAPPING OF HYDROGEN IN HIGH PURITY IRON A... (U) NORTHWESTERN UNIV EVANSTON IL DEPT OF MATERIALS SCIENCE AND E... J T WABER ET AL. 2/2

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7. AUTHOR(s) James T. Waber, Yong-Ki Park, C. Lewis Snead Jr.	8. CONTRACT OR GRANT NUMBER(s) DAAG 29 81-K-0153
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19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Positron Annihilation$_1$
Edge Dislocation$_2$
Screw Dislocation$_2$
Etch-Pit$_3$

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A strong 1 to 1 correlation is observed between the rate at which the numbers of etch pits and the number of traps increase with plastic deformation of the iron single crystals. A TEM investigation confirms these conclusions.

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DETERMINATION OF EDGE AND SCREW DISLOCATION DENSITY IN SINGLE CRYSTALS OF HIGH PURITY IRON

Yong-Ki Park,* James T. Waber,* and C. Lewis Snead Jr.,**

* Department of Materials Science and Engineering, Northwestern University, Evanston IL 60201

** Metallurgy and Material Sciences Division, Brookhaven National Lab, Upton, N. Y. 11973

The evidence for the trapping of positrons in two defects with lifetimes of 165 $\pm$ 2 and 142 $\pm$ 5 psec, in addition to 114 psec for annihilation in the bulk is presented together with the fitting parameters.

The general consensus is that the low binding energy of a positron to a dislocation (ca 0.1 eV) is insufficient to trap one and the positron migrates to a deeper trap before it annihilates. A jog is suggested as the trap and it should have a lifetime very sinmilar to that of a vacancy. Further, that there is too little room at the core of a screw dislocation to trap a positron.

However, for the following reasons, the authors believe that positrons are trapped by both edge and screw dislocation in B. C. C. metals. The fraction of positrons annihilating in both traps increases linearly with strain as the single crystal is deformed less than 10 percent. This fact establishes that these traps are associated with the generation of dislocations or at the very least, are derived from them.

Experiments designed to produce abundant screw dislocations, namely tensile straining at 200 K and twisting of iron whiskers, show that the fraction annihilating in the 142 trap increases with strain more rapidly than the fraction for 165 psec. One specimen observed to have a considerable amount of double slip - which should produce jogs - did not show strong evidence for 175 psec. The energy reported by others is 0.55 eV for the binding energy of a positron to a dislocation.

A more direct proof is the strong 1 to 1 correlation between the rate at which the numbers of etch pits and the number of traps increase with plastic deformation of the iron single crystals. A TEM investigation confirms these conclusions.

Research supported in part by the U. S. Army Research Office, DAAG 29 81-K-0153 and DUE Div. of Mat. Sci., Office of Basic Energy Sciences, under contract DE-AC 02-76-CH 00016

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Hydrogen Charging$_2$
Positron Traps$_2$
Hydrogen Traps$_2$
Hydrogen Occupation of Traps$_2$
Dislocation Trapping$_2$
Iron Single Crystal$_2$
Positron Annihilation$_3$
Hydrogen Embrittlement$_3$
Hydrogen Permeation$_3$

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The percentage of positrons annihilating in dislocation trap was observed to be reduced by the diffusion of hydrogen into the iron single crystals. The reduction was roughly 15 per cent for edge and nearly 50 per cent for screw dislocations. The former reduction is quite similar to the estimated occupancy of trapping sites along edge dislocation, namely 25%, which in turn is similar to the occupancy found from internal friction.

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REDUCTION OF THE TRAPPING OF POSITRONS IN DISLOCATED SINGLE CRYSTALS OF IRON WHEN CHARGED WITH HYDROGEN

Yong-Ki Park,* James T. Waber* and C. Lewis Snead, Jr.**

* Department of Materials Science and Engineering, Northwestern University, Evanston, Il 60201

** Metallurgy and Material Sciences Division, Brookhaven National Lab, Upton, N. Y. 11973

The percentages of the positrons annihilating in the two dislocation traps are diminished when approximately 1 atomic ppm of hydrogen is electrolytically charged into high purity single crystals of iron. Using the method of Devanathan and Stachurski[1] the cathodic and anodic current are monitored. The anodic side of the cell is equipped with a Coulometer and the decay current at constant voltage after the charging current has been turned off is recorded. Analysis of the permeation current yields the number of saturable and unsaturable traps. The former are in good agreement with the number of dislocations found from the lifetime spectra and Doppler line shape analyses. These traps increase monotonically with the strain of the specimens.

The number of edge and screw dislocations can be separated by means of the fraction of positrons annihilating in the 165 and the 142 psec traps. The influence of hydrogen "adsorbed" in the two types of traps can be assessed. There is no direct influence on the bulk lifetime - however, the fraction of positrons annihilating in the bulk increases when the other "channels" are blocked by hydrogen. It is found that with about 1 ppm of hydrogen present, the fraction annihilating in the screw component is decreased by nearly 40 percent whereas those annihilating in the edge dislocation traps is decreased by only about 15 percent.

These data permit one to estimate that at such low concentrations, the hydrogen concentration is small in the bulk and that only one in four (or five) sites along the edge dislocation are occupied. This figure is excellent agreement with the estimate Gibala[2] made on the basis of internal friction data.

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1. M. A. V. Devanathan and Z. Stachurski, Proc. Roy. Soc. (London) A270 90-102 (1962)

2 R. Gibala, Trans. Am. Inst. Met. Eng., 219 1575 (1967)

Research supported in part by U. S. Army Research Office, DAAG 29 81-K-0153 and by DOE Materials Science Div, Office of Basic Energy Sciences under contract DE-AC 02-76-CH 0000-16

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Lifetime in Screw Dislocations$_2$
Lifetime in Edge Dislocation$_2$
Etch Pit Measurements$_2$
TEM Examination$_2$
Positron Annihilation$_3$
Iron Single Crystals$_3$

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Positron annihilation measurements coupled with hydrogen permeation currents have permitted us to determine the number of dislocation traps and other defects in deformed single crystals of purified iron, and how the traps are occupied by hydrogen. Experiments dealing with hydrogen profiling at low hydrogen contents using ^{15}N and ^{3}He beams will be discussed. Embrittlement mechanisms will be reviewed.

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STUDY OF DISLOCATIONS AND HYDROGEN EMBRITTLEMENT OF IRON SINGLE CRYSTALS

by

James T. Waber, Yong-Ki Park and Wilfred Alsem

Materials Science and Engineering Department,
Northwestern University, Evanston, IL 60201

William Lanford

State University of New York at Albany,
Albany, N. Y. 12222,

C. Louis Snead Jr. and Kelvin G. Lynn

Brookhaven National laboratory, Upton, N. Y. 11973

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7. AUTHOR(s) A. Barry Kunz, Asok K. Ray, Robert Wiedman, James T. Waber		8. CONTRACT OR GRANT NUMBER(s) DAAG 29 81-K-0153
9. PERFORMING ORGANIZATION NAME AND ADDRESS Michigan Technological University Physics Dept. Houghton MI 49931		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
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Positron Attachment$_2$
Many-Body Perturbation Theory$_2$
Hartree-Fock$_2$
Positron
Correlation$_3$
Annihilation$_1$

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Using the method of Second Order, Many Body Perturbation Theory, a positron has been shown to bind to a variety of Closed Shell, Neutral atoms, even though it would not bind to them in the Hartree-Fock limit. Comparisons with other positronic atoms are discussed.

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MOLECULES INCORPORATING A POSITRON
II. - POSITRON ATTACHMENT TO NEUTRAL ALKALINE EARTH ATOMS AND ISOELECTRONIC ANIONS - AN ASSESSMENT OF CORRELATION EFFECTS

by

A. BARRY KUNZ

Physics Department,
University of Illinois at Champaign-Urbana
Champaign-Urbana, Il 61801

ASOK K. RAY*

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Physics Department,
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Houghton, MI 49931

and

JAMES T. WABER
Department of Materials Science and Engineering,
Northwestern University
Evanston, Il 60201

ABSTRACT

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Research Supported in part by the U. S. Navy ONR N 00014-18-K- 0020, by the U.S. Army Research Office, DAAG 29 81-K-0153 and U.S. National Science Foundation grant, PRM 8111589

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Electrons$_1$	Fermions$_2$	Positron Attachment$_3$
Negative Ions$_1$	Many-Body Perturbation Theory$_2$	Positrons$_3$
		Correlation$_3$

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The attachment of fermions which are distinguishable from the electrons to form exotic molecules has permitted us to eliminate the exchange potential and to concentrate on the correlation energy as a source of binding where there would be little if any tendency for electrostatic potential such as for a neutral atom to attract a particle. In contrast, a particle such as a positron or a muon would be strongly bound to an isoelectronic negative ion. The results obtained with positrons and muons, which differ essentially by their mass, permit us to look how this particle s wave function overlaps with those of the electrons and how it is affected by the particle mass.

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ABSTRACT

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CORRELATION IN EXOTIC MOLECULES
USING SECOND-ORDER MANY-BODY PERTURBATION THEORY FOR ASSESSMENT

by

James T. Waber
Northwestern University, Evanston IL 60201

Asok Ray
University of Texas at Arlington, Arlington TX 76019

A. Barry Kunz
University of Illinois at Urbana-Champaign IL 61801

and

Robert Weidman
Michigan Technological University, Houghton MI 49931

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The results obtained with positrons and muons, which differ essentially by their mass, permits us to look how this particle's wave function overlaps with those of the electrons and how it is affected by the particle mass.

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Fermion Mass$_2$
Bound States$_2$
Anions$_2$
Positron Attachments$_3$
Correlation$_3$
Positron Attachments$_2$

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The effect of the mass of a series of positively charged Fermions on the formation of bound states with closed shell anions will be discussed. The fairly rich but contracted basis for the anions was augmented by several diffuse orbital plus bond polarization functions. The Fermion-electron correlation energy was estimated using Second Order, Many-Body Perturbation Theory.

Presented at 8th Canadian Conference on Theoretical Chemistry, August 1983

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EFFECT OF FERMION MASS ON THE FORMATION OF BOUND STATES IN HYDRIDE-LIKE EXOTIC SPECIES.

by

JAMES T. WABER
Department of Materials Science and Engineering,
Northwestern University
Evanston, IL 60201

NEIL ROETH
Physics Department,
Colgate University,
Hamilton, NY 13346

and

A. BARRY KUNZ
Physics Department,
University of Illinois at Champaign-Urbana
Champaign-Urbana, IL 61801

ABSTRACT

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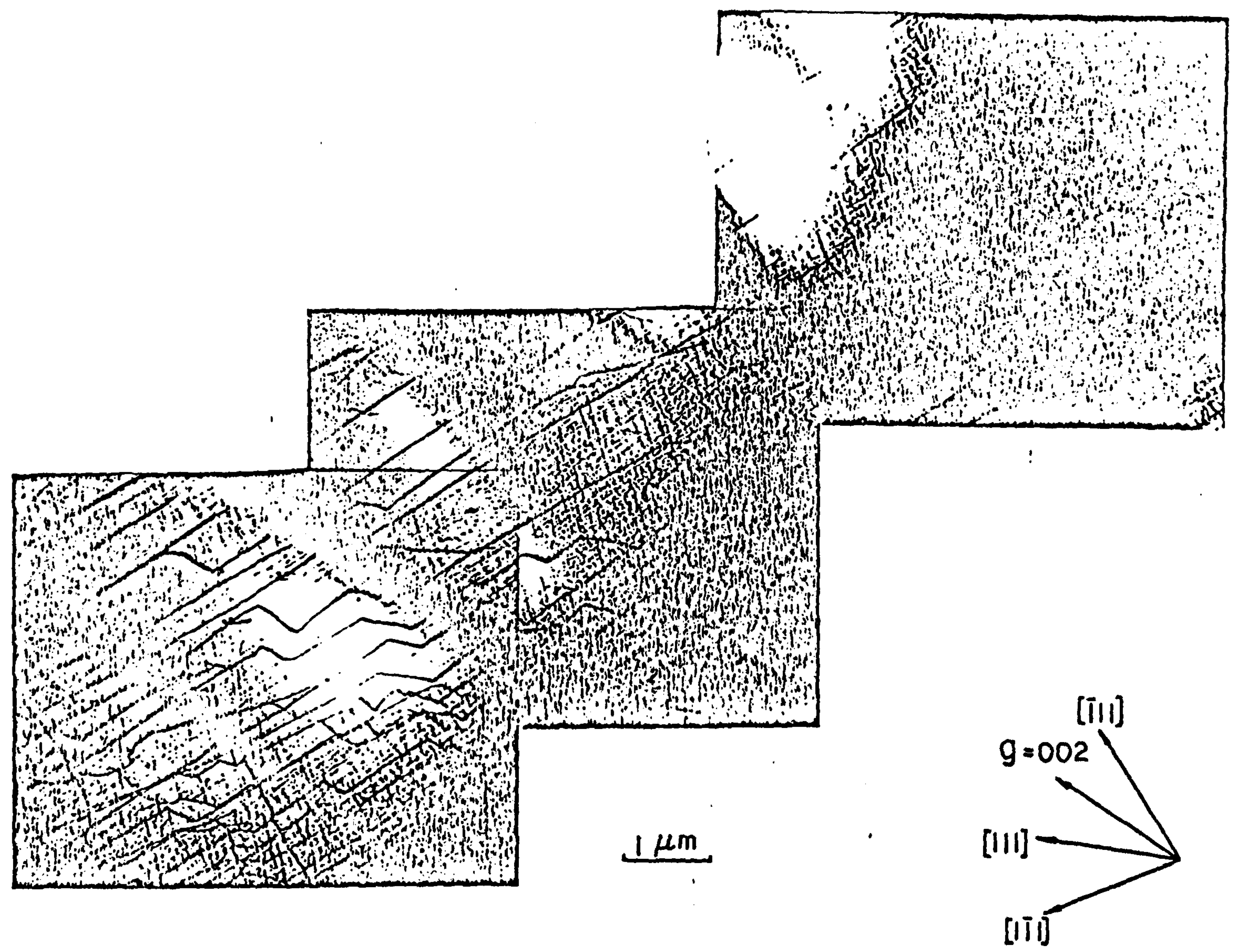
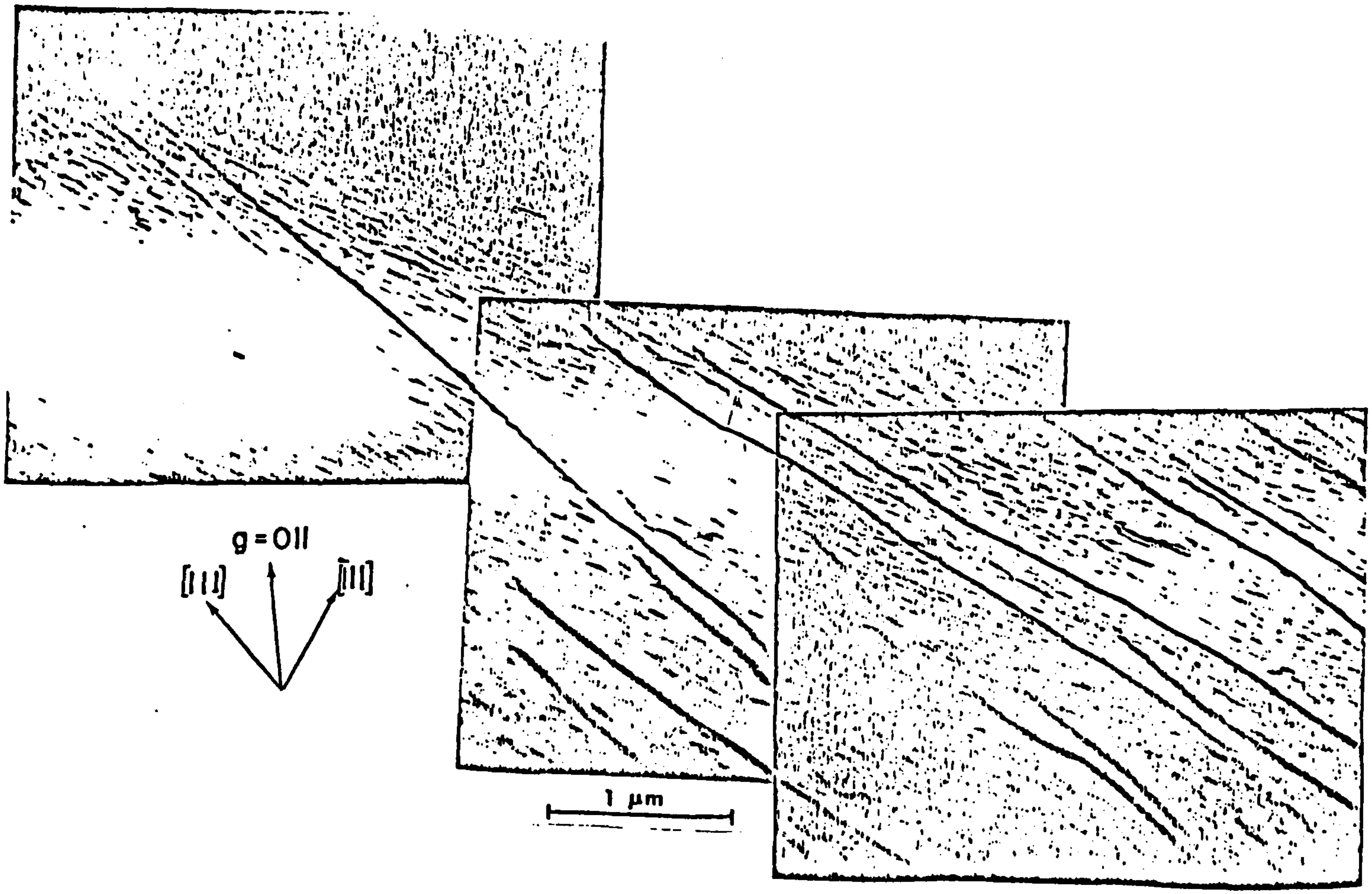
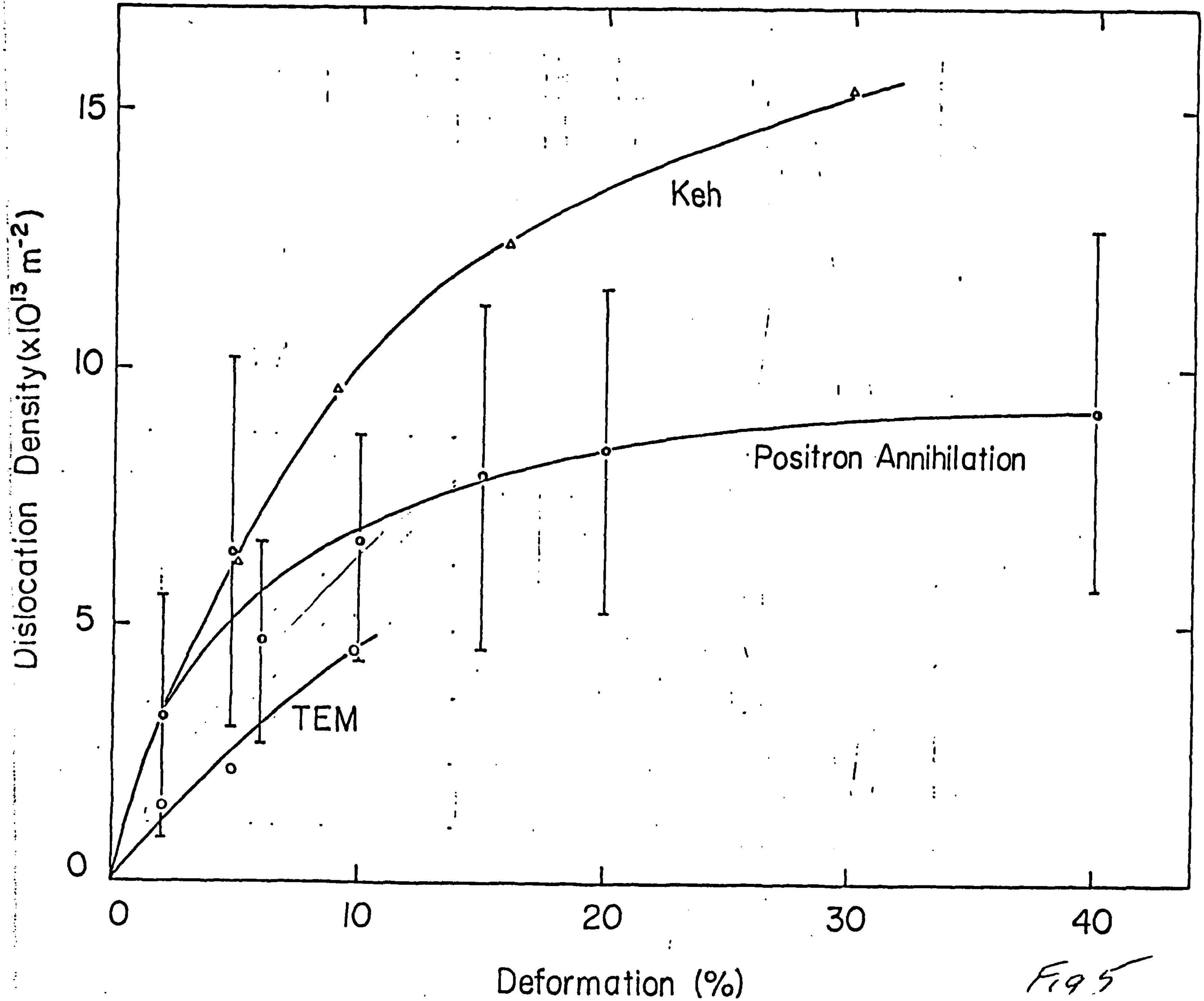


1 μm
[$\bar{1}$11]
g=002
[111]
[1$\bar{1}$1]

Fig 4





Dislocation Density ($\times 10^{13}$ m^{-2})
15
10
5
0
Keh
Positron Annihilation
TEM
0
10
20
30
40
Deformation (%)

Fig 5

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Lifetime in Edge Dislocation$_2$
Etch Pit Measurements$_2$
TEM Examination$_2$
Positron Annihilation$_3$
Iron Single Crystals$_3$

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The trapping of positrons in dislocation-associated traps has been studied and the density of traps has been demonstrated to be in close agreement with the density of dislocations determined by TEM and etch-pit measurements on the same specimens. The specific trapping rates were determined.

Accepted for Indian Conference on Positron Annihilation, New Delhi, Jan. 1985.

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DETERMINATION OF EDGE AND SCREW DISLOCATION DENSITY IN SINGLE CRYSTALS OF HIGH-PURITY IRON

Yong-Ki Park,* James T. Waber,* and C. L. Snead, Jr.,**

* Department of Materials Science and Engineering, Northwestern University, Evanston IL 60201

** Metallurgy and Material Sciences Division, Brookhaven National Lab, Upton, N. Y. 11973

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INTRODUCTION

Dislocations have been recognized as traps for positrons for some time[1], but little specific research has been done to deduce the type of dislocation involved, or the specific trapping rate.

The general consensus has been that the low binding energy of a positron to a dislocation (ca 0.1 eV) is insufficient to trap one and the positron *migrates to a deeper* trap before it annihilates.[2,3] A jog is suggested as the trap and it should have a lifetime very similar to that of a vacancy[3,4]. Further, it has been believed that there is too little room at the core of a screw dislocation to trap a positron.

However, the authors present the evidence herein that positrons are trapped by both screw and edge dislocations in B.C.C. iron. The fraction of positrons annihilating in both traps, namely those resp., with a lifetime of 142 ± 5 psec and those with the 165 ± 3 psec are oberved to increase montonically with strain as the single crystal is deformed. This fact establishes that these traps are associated with the generation of dislocations or at the very least, with some species derived from them. Deformations have been limited in most cases to less than 10 percent to minimizes dislocation interactions.

In the present work, single crystals were deformed in three different ways, namely to produce (a) almost exclusively edge dislocations by bending, (b) abundant screw dislocations, namely by tensile stretching at 200 K and (c) a mixture of edge and screw by cold rolling at room temperature. Both Doppler broadening and positron lifetime measurements were made and the data analyzed.

DATA ANALYSES

The lifetimes determined for the bent specimen were 114 psec for the trap-free bulk and 165 psec for the edge dislocation. The number of redundant dislocations were larger on the convex side that on the concave side. No evidence for 142 psec was obtained.

On the basis statements in the literature[5], the fraction of screw dislocations increase with strain at 200 K more rapidly than the fraction of edge dislocations. We observed as expected, that the fraction of positrons annihilating in the 142 trap increased with strain more rapidly than the fraction for 165 psec. This gave considerable support to associating the 142 psec trap with annihilation in a screw dislocation.

The third type of specimen namely, cold-rolled single crystals, was used to obtain a preliminary figure for the "average" specific trapping rate namely 10×10^{-5} m/sec. The resulting planar density of dislocations were found to be in good agreement with the TEM measurements of Keh[6]. In subsequent experiments, trapping rates κ_T for both edge and screw dislocations were obtained by fitting our positron data with a two-trap model. The relative

proportion of edge and screw components formed during deformation were assumed to be the same as in the experiments of Yamakawa *et al.*[7] who studied low carbon (0.06%) polycrystalline samples of iron using Xray diffraction and hydrogen permeation methods; then he compared his data with published TEM data.

With such data in hand, the specific trapping rates were found to be 7×10^{-8} for edge dislocations and 5×10^{-8} m^2/sec for screw dislocations. These values are in reasonable agreement with the estimate of Cao *et al.*[8]. The planar density of screw and edge dislocations were deduced from these rates.

Two more direct determinations were made. The density of etch pits and the TEM density of dislocations were compared with the densities of positron trpas.

EXPERIMENTAL RESULTS

The details of the experimental procedures were discussed elsewhere recently[9]. One of TEM photo-micrographs of dislocations is shown in *Figures 1. This identifies the* dislocations formed at 200 K as being primarily of the <111> type of screw dislocation.

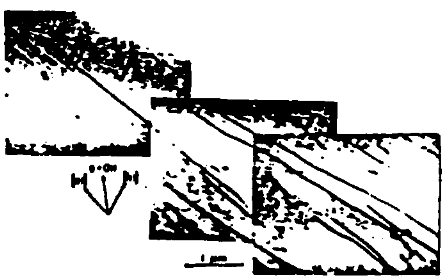

Figure 1. Dislocation on (011) planes of crystal elongated 9.8% at 200 K. Long screw dislocations are present.

In addition, densities of dislocations in cold rolled specimens were estimated from a number of TEM photographs.

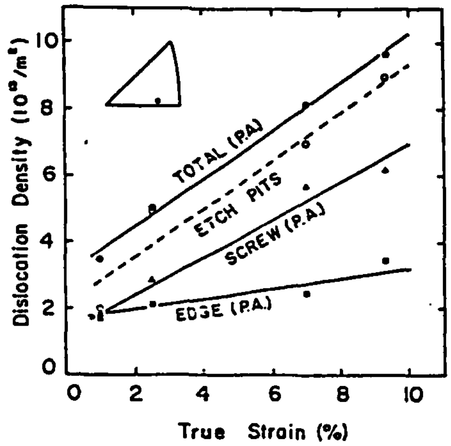

Figure 2. Comparison of dislocation densities measured by positron annihilation and etch pits in iron *deformed in tension at 200 K.*

COMPARISON OF RESULTS

The strong 1-to-1 correlation between the rate at which the numbers of etch pits and the number of traps increase with plastic *deformation of the same iron single* crystals is evident in Figure 2. In addition, the density of dislocations determined by TEM and positron annihilation are compared in Figure 3. The TEM results of other authors[6,10,11] are plotted in this figure. The TEM, the etch pits, and the positron lifetime results depend on plastic deformation in very parallel ways.

These dependences taken together offer strong evidence for positron annihlation in the dislocations.

However, the question of whether annihilation occurs in jogs can not be settled by the present experiments. The number of possible traps for positrons per cubic centimeter can be calculated from the product of (a) the planar density of dislocations and (b) the number of traps per unit length. The latter is based on the assumption similar to that of Gibala[12] that one trap might occur every lattice parameter distance along the dislocation line. A typical number of traps is 10 or less per million lattice sites.

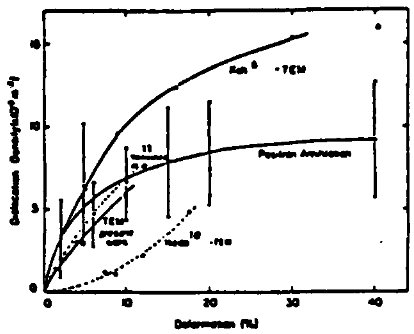

Figure 3. Comparison of dislocation density measured by TEM and positron annihilation in cold rolled specimens. The TEM results of Keh[6], Ikeda[10] and Yamashita *et al.*[11] are included for comparison.

It has been proposed that the separation between jogs might be 100 to 1000 Burgers' vectors.[2] The separation of jogs in the present crystals can be estimated from the TEM photographs to be approx. 2000 Burgers' vectors. The number of this type of jog which might act as a trap is at least 3 orders of magnitude less than the density of positron traps. Recently, Weertman[13] pointed out that the jogs visible in these photographs are large jogs and that there may in addition be many jogs of almost atomic dimension which can not be easily resolved. While there are insufficient large-jogs to account for the annihilation, the unresolved (small) jogs can not be ruled out as being the sites for positron annihilation.

CONCLUSIONS

The connection between positron annihilation traps and dislocations of both the screw and edge types has been established. Two lifetimes have been observed, 142 ± 5 for screw and 165 ± 3 psec for edge components. Specific trapping rates have been determined. On the same single crystals, the density of *(i)* positron traps, *(ii)* of etch pits and *(iii)* of dislocations observed in TEM studies exhibit very similar dependences on the plastic deformation of the crystals. The possibility of trapping at jogs can not be ruled out.

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